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AFOSR-TR-75-0384 F49620-83-C-0133

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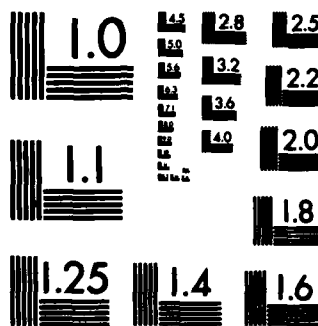
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RADIATIVE AUGMENTED COMBUSTION

MOSHE LAVID  
M. L. ENERGIA, INC.  
P.O. BOX 1468  
PRINCETON, NEW JERSEY 08542

AUGUST 1984

ANNUAL TECHNICAL REPORT AFOSR-TR-~~85-0384~~  
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## 19. ABSTRACT (continued)

effort. The objective of the experimental work is to demonstrate both ignition and enhancement in flow systems. For this purpose, two specially tailored apparatus were designed and fabricated; a plug flow combustor to conduct radiative ignition tests, and a "pancake" burner to measure combustion enhancement in terms of increase in burning velocity. It is expected that experiments will demonstrate proof of concept by igniting combustible mixtures at conditions where thermal ignition is unreliable, and by increasing the flame speed. Since the role of photochemical reactions within the whole kinetic scheme is currently not completely understood, it is further expected that this research will identify the most effective photodissociative paths. This, in turn, will assist in determining the feasibility of the concept based on the availability of newly developed light sources and their compatibility with the harsh combustion environment.

Consistent with the experimental work research in the area of analytical modeling has also proceeded on two fronts: ignition and enhancement. Ignition modeling revealed the inhibiting effect of  $\text{HO}_2$  and  $\text{H}_2\text{O}_2$  in hydrogen flames which retards the build-up of free radicals necessary for chain-branching ignition. The use of the HCT model to simulate radiative enhanced combustion has continued. The most significant finding was a substantial increase in burning velocity (28%) with the inclusion of the photodissociation of the metastable hydroperoxyl radical.

This result further supports the concept of radiative augmented combustion and points to new practical opportunities for combustion enhancement with relaxed light source and optical requirements. These are being pursued both experimentally and theoretically.

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MATTHEW J. KENNEDY  
Chief, Technical Information Division

FOREWARD

This is the first annual technical report on research on Radiative Augmented Combustion conducted at M. L. ENERGIA. It is funded by the Air Force Office of Scientific Research under Contract No. F49620-83-C-0133, with Dr. J.M. Tishkoff as the Program Manager. This research under a new contract succeeds and continues the work performed under the previous Contract No. F49620-81-C-0028 at Exxon Research and Engineering Co., Linden, New Jersey.

This report covers the research progress over the first year of the contract; since its inception on July 15, 1983 through July 14, 1984. The work was performed at ENERGIA, Princeton, New Jersey, with Dr. M. Lavid as the Principal Investigator.



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ABSTRACT

Radiative Augmented Combustion has been identified as a potential technique for extending aircraft operating limits associated with combustion processes. This technique is based on the fact that radiation of selected wavelengths is capable of photodissociating stable molecules, combustion intermediates and other inhibiting species in the flame zone into reactive radicals. Subsequent reactions involving these reactive radicals lead to radiative ignition and combustion enhancement. The potential of this radiative technique has been already demonstrated under laboratory static conditions. It has successfully ignited fuel-air mixtures, and has enhanced combustion processes utilizing vacuum ultraviolet and ultraviolet radiations emitted by high pressure xenon lamps and newly developed excimer lasers.

This program emphasizes research on the interaction between radiation and combustion under flow conditions. Research is divided into two main subjects; ignition and enhancement. Each subject consists of an experimental effort supplemented by an analytical effort. The objective of the experimental work is to demonstrate both ignition and enhancement in flow systems. For this purpose, two specially tailored apparatus were designed and fabricated; a plug flow combustor to conduct radiative ignition tests, and a "pancake" burner to measure combustion enhancement in terms of increase in burning velocity. It is expected that experiments will demonstrate proof of concept by igniting combustible mixtures at conditions where thermal ignition is unreliable, and by increasing the flame speed. Since the role of photochemical reactions within the whole kinetic scheme is currently not completely understood, it is further expected that this research will identify the most effective photodissociative paths. This, in turn, will assist in determining the feasibility of the concept based on the availability of newly developed light sources and their compatibility with the harsh combustion environment.

Consistent with the experimental work research in the area of analytical modeling has also proceeded on two fronts: ignition and enhancement. Ignition modeling revealed the inhibiting effect of  $\text{HO}_2$  and  $\text{H}_2\text{O}_2$  in hydrogen flames which retards the build-up of free radicals necessary for chain-branching ignition. The use of the HCT model to simulate radiative enhanced combustion has continued. The most significant finding was a substantial increase in burning velocity (28%) with the inclusion of the photodissociation of the metastable hydroperoxyl radical.

This result further supports the concept of radiative augmented combustion and points to new practical opportunities for combustion enhancement with relaxed light source and optical requirements. These are being pursued both experimentally and theoretically.

### STATEMENT OF WORK

The contractor shall furnish scientific effort together with all related services, facilities, supplies and materials needed to conduct the research described below. The research shall be conducted during the period July 15, 1983 through July 14, 1984. The professional and non-professional level of effort for this period is 1550 and 1160 man-hours, respectively.

- a. Provide a combustion test facility capable of conducting radiative ignition and enhancement experiments under flow conditions. Examine the possibility of using a vertical combustion chamber rather than present horizontal one to eliminate gravitational effects.
- b. Select and employ the most appropriate VUV/UV sources from the currently available devices; ILC, EIMAC, and Excimer lasers. Review and search literature for new developments in VUV/UV capabilities and incorporate into the program any improved capabilities that may become available.
- c. Design and conduct radiative ignition and enhancement tests under flow conditions. Investigate the effects of equivalence ratio, pressure and velocity (laminar and turbulent). Measure and compare results under static and flow conditions, and then compare with conventional spark ignition data. Measure enhancement in terms of flame propagation velocity and extinction limits and investigate the effects of exposure time and optical paths on combustion enhancement.
- d. Incorporate into the static initiation (ignition) model the convective heat and mass (species) transfer terms. Compare experimental data with model predictions. Undertake additional model revision and refinement as deemed necessary from the comparison.
- e. Modify and revise the HCT model to simulate radiative enhancement under static and flow conditions. Calculate flame propagation properties and compare with experimental data. Ascertain agreement between experimental and analytical results and resolve any discrepancies. Use the model to explore the interaction between spectral absorptance and subsequent photo and chemical kinetics, and transport phenomena in reacting flow systems. Utilize the analytical effort as an adjunct to the experimental work to provide guidance and ultimately to assist in optimization of the system performance.

f. Analyze and assess the experimental and analytical results to determine the feasibility of utilizing the radiative technique in practical combustion hardware. Develop a plan for larger scale combustion tests of the most promising radiative technique(s) and provide pertinent information on all aspects of the radiative technique(s).

### STATUS OF THE RESEARCH

This section describes the research progress since its inception on July 15, 1983, and covers the work performed through July 14, 1984. The program emphasizes radiative augmented combustion under flow conditions. It is divided into two parts: Ignition and enhancement. Each part contains an experimental effort supplemented by an analytical effort.

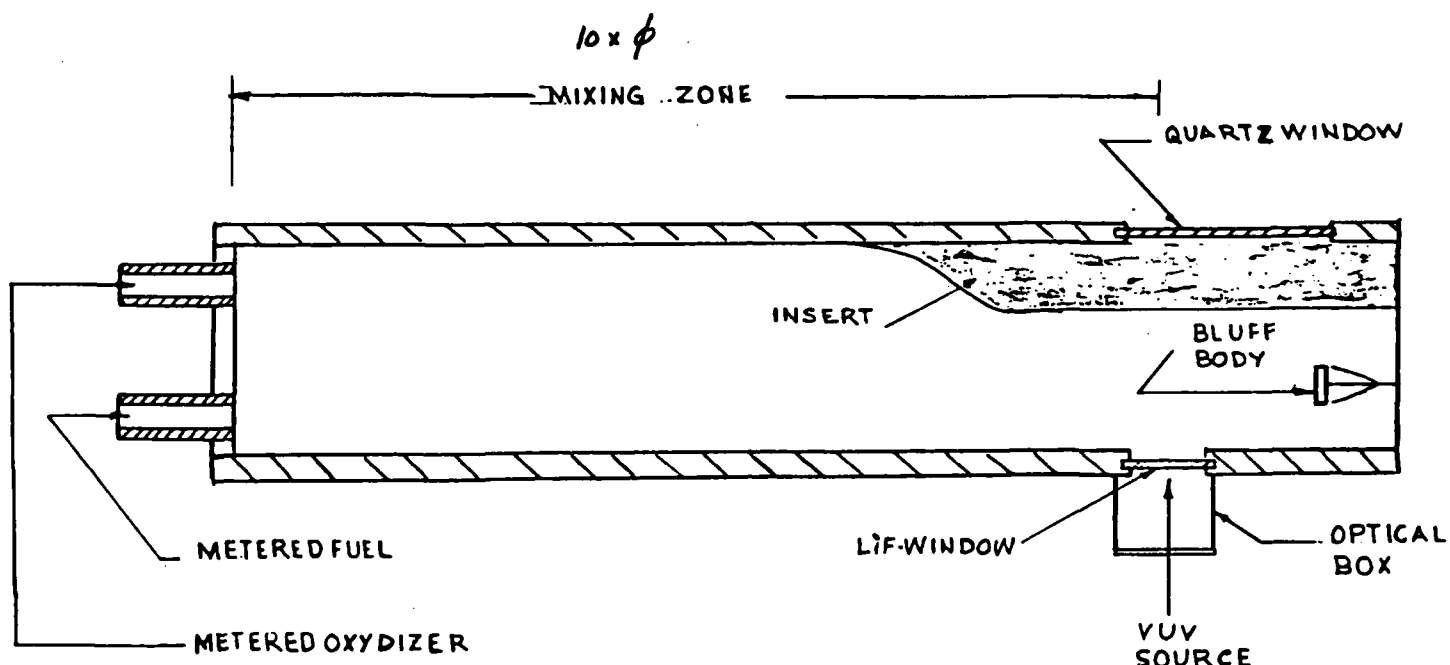
#### A. Experimental Effort

The transfer of the combustion equipment from the previous contractor was completed after a long delay. Although the original contract was already signed on November 7, 1983, the equipment was transferred only on March 9, 1984 because of legal issues raised by the lawyers of the previous contractor. Upon arrival, it was found that few items either disappeared or were damaged while being at idle storage. We decided to design and fabricate two new apparatus rather than repair or replace the original ones. These two apparatus are described below. Fortunately, VUV/UV light sources, the power suppliers and the other optical components were received in working conditions.

Ignition: The objective of this part is to obtain successful radiative ignition under flow conditions, and to characterize it in terms of minimum radiative ignition energy as a function of wavelength, equivalence ratio and velocity.

The design of the radiative ignition apparatus is completed. It is based upon a plug flow combustor shown in Figure 1. The combustor is capable of operating with premixed combustible gases at various equivalence ratios, at ambient temperature and atmospheric pressure. The velocity of the incoming gas mixture can be varied by changing the mass flows and/or by employing inserts which change the cross-sectional area at the test section. An optical box generates, conditions, and delivers VUV irradiation to the ignition site (nearby the window).

Because the radiative input is pulsed, flames which are ignited at the window will be swept downstream. To stabilize these flames within the test section, a bluff body is provided downstream of the window. This unique design ascertains low temperature in the vicinity of the optical box, protecting the



## SECTION AREAS

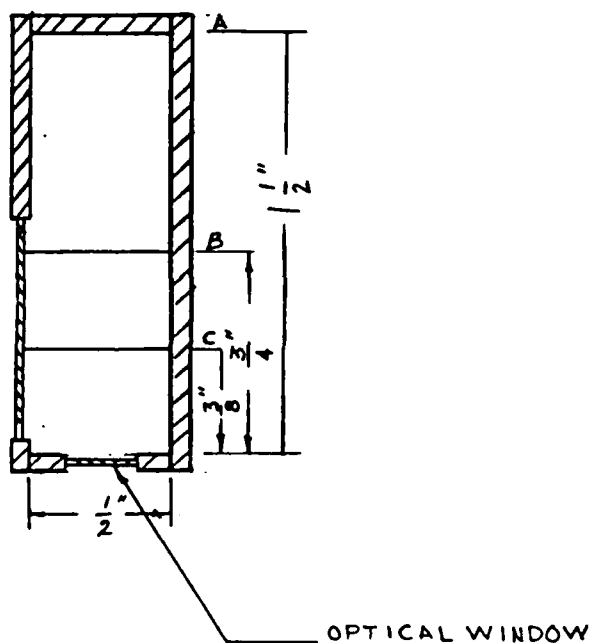
A - 0.75 in<sup>2</sup>B - 0.375 in<sup>2</sup>C - 0.1875 in<sup>2</sup>QUARTZ  
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FIGURE 1  
RADIATIVE IGNITION APPARATUS

window from high flame temperatures. Ignition tests conducted on this plug flow combustor will provide data for minimum radiative ignition energy as function of wavelength, equivalence ratio and velocity. During the first phase, the velocity will be maintained in the laminar region, where flame theory is better understood. Tests will begin with hydrogen flames and later will proceed with propane flames. The light sources to be employed are ILC and Excimer Laser ( $F_2$ ).

The main advantage of laser over short arc plasma lamps is in its well characterized emission. A lamp has a wide spectrum of output with only a small fraction (less than 5%) in the VUV/UV wavelength region (ENG 102, REVISED, p. 33). Conversely, a laser output is at a discrete and very narrow wavelength band, which makes it an excellent research tool, provided, of course, an "excimer" molecule (rare gas halide) does exist in the VUV region and emits adequate pulse energy. Fortunately, with the recent and rapid development in laser technology, such lasers are now commercially available.

Earlier laser ignition experiments at Los Alamos using Lumonics TE 861 excimer lasers demonstrated both the usefulness of these lasers to this work and the importance of VUV spectral distribution to the success of the experiments. The excimer lasers were employed at two VUV wavelengths; 157 nm ( $F_2$ ) and 193 nm (ArF). Radiative ignitions were obtained with the fluorine laser and not with the argon fluoride laser, even though the latter has about twenty fold greater pulse energy. This was a convincing demonstration of the importance of spectral selectivity to the radiative augmented combustion research.

The employment of excimer lasers in the ignition tests under flow conditions does not present any significant problem. In essence, they will be used in the same manner as the ILC lamps.

Enhancement: The objective of this part of the research program is to detect any radiative combustion enhancement under flow conditions. After considerable technical discussions, summarized below, we decided to measure enhancement in terms of changes in laminar burning velocity.

1. **DISCUSSION OF THE PROBLEM:** The concept of laminar burning velocity unambiguously concerns only one-dimensional flames in laminar streams of premixed combustible gases. Departures from one-dimensionality are tolerable if the radius of curvature of the flame front is very much larger than the flame zone thickness. Also, departures from strict pre-mixedness are tolerable if, in the vicinity of the flame zone, mass diffusion rates greatly exceed chemical kinetic rates, in which case the chemical rates are controlling. If these conditions are met, the concept of laminar flame velocity is applicable to the experiment.

Assuming that a laminar flame front exists in the experimental apparatus, it is necessary to deliver ultraviolet radiation to the immediate vicinity of the reaction zone if it is to affect the flame propagation velocity. Also, if the results of the experiment are to be amenable to interpretation in one-dimensional terms, this radiation must be deposited in a reasonably uniform density over the area of the flame that is being studied; and it must produce effects on the flame that are measurable and which can be related to the laminar burning velocity. These are difficult conditions to achieve in experimental apparatus.

As an example, consider the ideal case of an infinite flame in the YZ plane which is fixed at an X-axial point (laboratory coordinates), in a uniform laminar premixed hydrogen/oxygen stream, flowing in the positive X-direction. The velocity of this cold stream,  $U_0$ , is by definition the laminar flame velocity. Upstream from the flame front, the mixture consists of the reactants  $H_2$  and  $O_2$ . Downstream from the flame front, it consists of the products  $H_2O$ ,  $H_2$ ,  $O_2$ , and minor free radical species. In an over (under) oxidized stream,  $H_2$  ( $O_2$ ) will be largely absent in the product gas.

Suppose now that the flame front is illuminated from the reactants side by UV radiation propagating in the positive X-direction. This radiation, which is chosen to interact strongly with  $O_2$ , will be absorbed by the  $O_2$  in the reactants and only part of it will reach the flame front. (The  $H_2$  in the reactants will not absorb UV radiation in the wavelength range that will be used.) If the



optical pathlength through the reactants is long relative to the flame zone thickness, far more radiation will be deposited remote from the flame zone than in it. If the radiation is strongly absorbed along this path, the UV radiative flux reaching the flame front will be quite low unless the input flux is extremely high. The flux deposited in the reactants will dissociate  $O_2$  to  $O$  which will then either react with the  $H_2$  or will recombine to  $O_2$  with attendant dissipation of the absorbed energy. Unless the reaction with  $H_2$  is sufficiently vigorous to result in radiative ignition, upstream from the flame, the UV absorbed in the reactants far from the flame zone will not influence the flame propagation.

Alternatively, the radiation may be introduced from the product side. If the products are transparent to the UV radiation, it will reach the flame zone undiminished. Unfortunately, the UV absorption bands of  $O_2$  and  $H_2O$  overlap, and while  $H_2O$  absorbs less strongly than  $O_2$ , its absorption is still significant. The lower absorption of the products does favor introducing the radiation from the products side. However, if optical pathlengths do not greatly exceed those that may be used from the reactants side, the practical experimental problem of protecting UV-transparent windows from hot combustion products is likely to require substantial pathlengths, and it argues in favor of introducing the radiation from the reactants side. From either side, it is necessary to provide short pathlengths and low densities of absorbers.

A real laboratory flame is not of infinite extent in the YZ plane. Small flames with low densities of absorbers may be illuminated from the side and still be relatively uniformly illuminated across their surface if the radiation is only slightly absorbed during its entire pathlength. If conditions exist, side illumination is a third experimental possibility.

2. DISCUSSION OF THE MEASUREMENT PROBLEM: To simplify the experimental procedure, only flames fixed in laboratory coordinates will be considered. Specifically, only flat flames (porous burner flames) or conical flames (bunsen flames) will be considered.

In the case of flat flames, adiabatic burning velocities may be determined by the method of Spalding (1). In the case of conical flames, velocity determinations may be made from images (either luminous, shadow, schlieren, or interferometric) of the flame cone (2).

We wish to determine the departure of the burning velocities of UV illuminated flames from those of non-illuminated flames. Thus, we will perform differential experiments in which experimental flames are periodically illuminated with UV radiation and their burning rates, with and without illumination compared. The following examples show how this may be done for these two types of flames.

Figure 2 shows a schematic graph of flame speed,  $U$ , versus heat loss,  $Q$ , for a typical Spalding flat flame burner. Curves A and B represent hypothetical operating lines for non-illuminated and illuminated flames, respectively. Over a large portion of their range, below some high value of  $Q$ , these curves are linear. At very low  $Q$ , the flames become unstable. The adiabatic burning velocity,  $U_0$ , is obtained by extrapolating these curves to  $Q = 0$ . The equations for the linear ranges of these curves are:

$$U_A = U_{OA} + \tan \phi_A Q_A \quad (1)$$

$$U_B = U_{OB} + \tan \phi_B Q_B \quad (2)$$

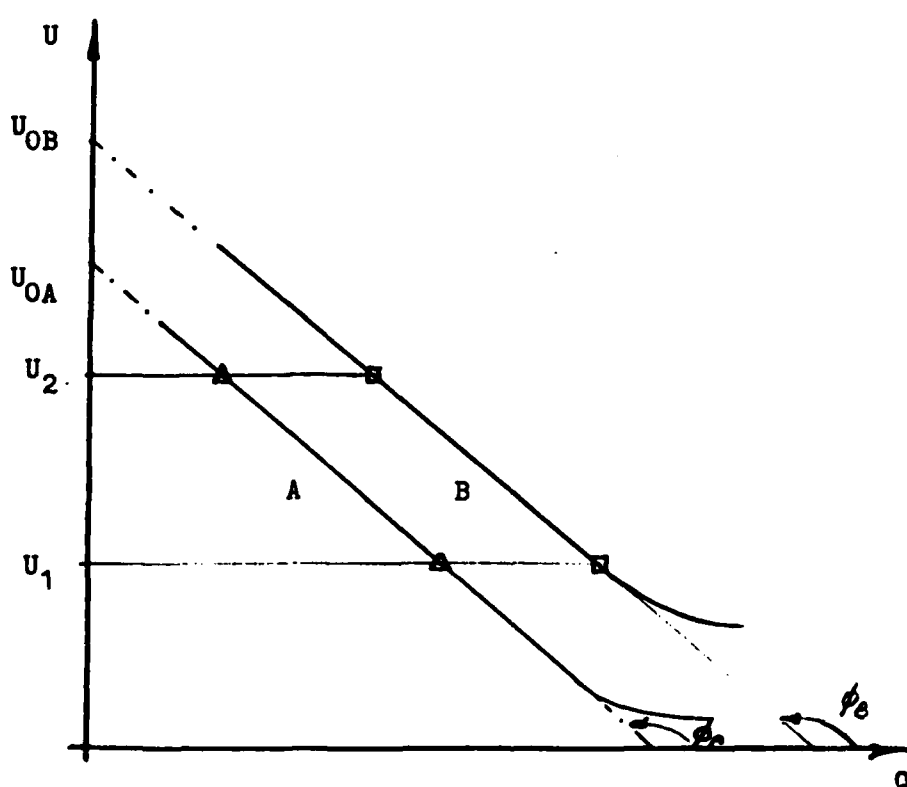


FIGURE 2

SCHEMATIC GRAPH OF OPERATING LINES OF A LAMINAR FLAT FLAME BURNER WITH CURVE A REPRESENTING NON-ILLUMINATED AND CURVE B REPRESENTING ILLUMINATED FLAMES

Suppose now that two differential experiments are run in the linear ranges of A and B; one at flow velocity  $U_1$ , and one at flow velocity  $U_2$ . For each experiment,  $Q$  is measured alternately in the non-illuminated and the illuminated states. These measurements give rise to four equations .

$$U_1 = U_{OA} + \tan\phi_A Q_{A1} \quad (3a)$$

$$U_1 = U_{OB} + \tan\phi_B Q_{B1} \quad (3b)$$

$$U_2 = U_{OA} + \tan\phi_A Q_{A2} \quad (3c)$$

$$U_2 = U_{OB} + \tan\phi_B Q_{B2} \quad (3d)$$

Taking differences gives:  $U_2 - U_1 = \tan\phi_B (Q_{B2} - Q_{B1}) = \tan\phi_A (Q_{A2} - Q_{A1})$

$$\tan\phi_B = \tan\phi_A (Q_{A2} - Q_{A1}) / (Q_{B2} - Q_{B1}) \quad (4)$$

Also, from equations 3, one obtains:  $U_{OB} - U_{OA} = (\tan\phi_A) Q_A - (\tan\phi_B) Q_B$

$$(U_{OB}/U_{OA}) - 1 = (\tan\phi_A Q_A - \tan\phi_B Q_B) / U_{OA} \quad (5)$$

where  $Q_A$  and  $Q_B$  are measured at a common flame speed  $U = U_1$ , or  $U = U_2$ .

Suppose now that the non-illuminated characteristic Curve A has been determined experimentally. Then with equations 4 and 5, the fractional change in  $U_0$  that is caused by a given intensity of illumination can be determined from the results of only two differential experiments.

As the second example, consider a conical bunsen flame with burning velocity  $U_0$  that is stabilized in a uniform gas jet with axial flow velocity  $U > U_0$ . The idealized picture of this system is shown in Figure 3.

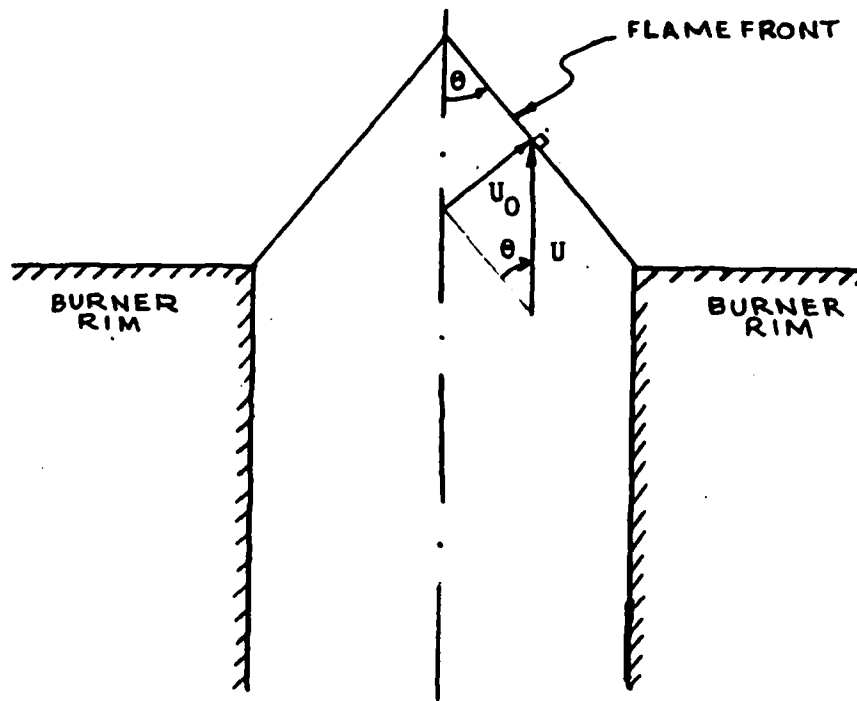


FIGURE 3

SCHEMATIC DIAGRAM OF BUNSEN FLAME WITH BURNING VELOCITY  $U_0$  STABILIZED IN UNIFORM GAS STREAM FLOWING WITH AXIAL VELOCITY  $U > U_0$ .

The vector relationship gives, in terms of velocity magnitudes:

$$U_0 = U \sin \theta \quad (6)$$

If again, the non-illuminated flame is labeled A and the illuminated flame is labeled B, then:

$$U_{OB} - U_{OA} = U (\sin \theta_B - \sin \theta_A) \quad (7)$$

Rewriting equation 6 as:

$$\theta = \sin^{-1} (U_0 / U) \quad (8)$$

and differentiating and rearranging:

$$\frac{d\theta}{dU_0} = 1 / (U_0 \sqrt{(U / U_0)^2 - 1}) \quad (9)$$

Equation 9 shows that the sensitivity of  $\theta$  to changes in  $U_0$  increases as  $U$  approaches  $U_0$ , i.e., as the flame approaches a flat plane perpendicular to the flow.

If the UV induced changes in the burning velocity are very small, they may be difficult to detect by either of the above two methods. Then, with either the flat flame or the bunsen burner, a method of acoustic detection may prove to be considerably more sensitive. The case of the flat flame burner provides a simple example. Assume that a flat flame is burning in the non-illuminated state A, with burning velocity  $U_{A1}$ , and that it is suddenly illuminated. The burning velocity increases to  $U_{B1}$  with  $Q$  remaining as  $Q_1$ . At this point,  $U_{B1} > U$ , the stream velocity, and the flame propagates upstream a distance  $\delta X$  to a new equilibrium position, which results in a higher heat loss,  $Q_2$ , and a new flame velocity,  $U_{B2} = U$ . If  $S$  is the affected area of the flame, this movement consumes a volume of reactants  $S \cdot \delta X$  in excess of that which it would have consumed had it not been illuminated. This will result in an acoustic pressure pulse. Similarly, cessation of illumination will result in a rarefaction pulse. Thus, if the burning velocity is affected by illumination, chopping the input radiation will result in an acoustic signal. The detection of this signal will constitute an extremely sensitive method of detecting changes in the burning velocity.

3. THE INITIAL EXPERIMENTAL BURNER: The first burner that is constructed for this work will produce a conical bunsen-type flame. This burner was chosen because of the experimental simplifications it offers in achieving axisymmetric illumination with short optical pathlengths. It also has the window on the cold reactant side of the flame where it is not subjected to high temperature.

A schematic diagram of the burner is shown in Figure 4. The reactive gas exits as a jet from a small orifice in the thin top plate of a plenum chamber. The conical flame burns in this jet and is anchored at the circumference of the orifice. The plenum chamber, which is fed with premixed gaseous reactants, is a thin pancake only one to three millimeters thick. The orifice diameter is of the order of two millimeters. The optical pathlength from window to flame front will be in the range of two to four millimeters. The optical density may be reduced by operating at subatmospheric pressure. Detailed calculations to obtain conditions for maximum UV at the flame zone are given in Appendix A.

The reactants, initially, will be hydrogen and air. Either the shadow or schlieren images of the flame will be used to determine the burning velocity. Acoustic detection will also be tested. The details of the burner and the

detection system are presently being designed.

This apparatus will provide for efficient and economical first experiments on the effects of ultraviolet radiation on the fundamental burning velocity of hydrogen/air flames.

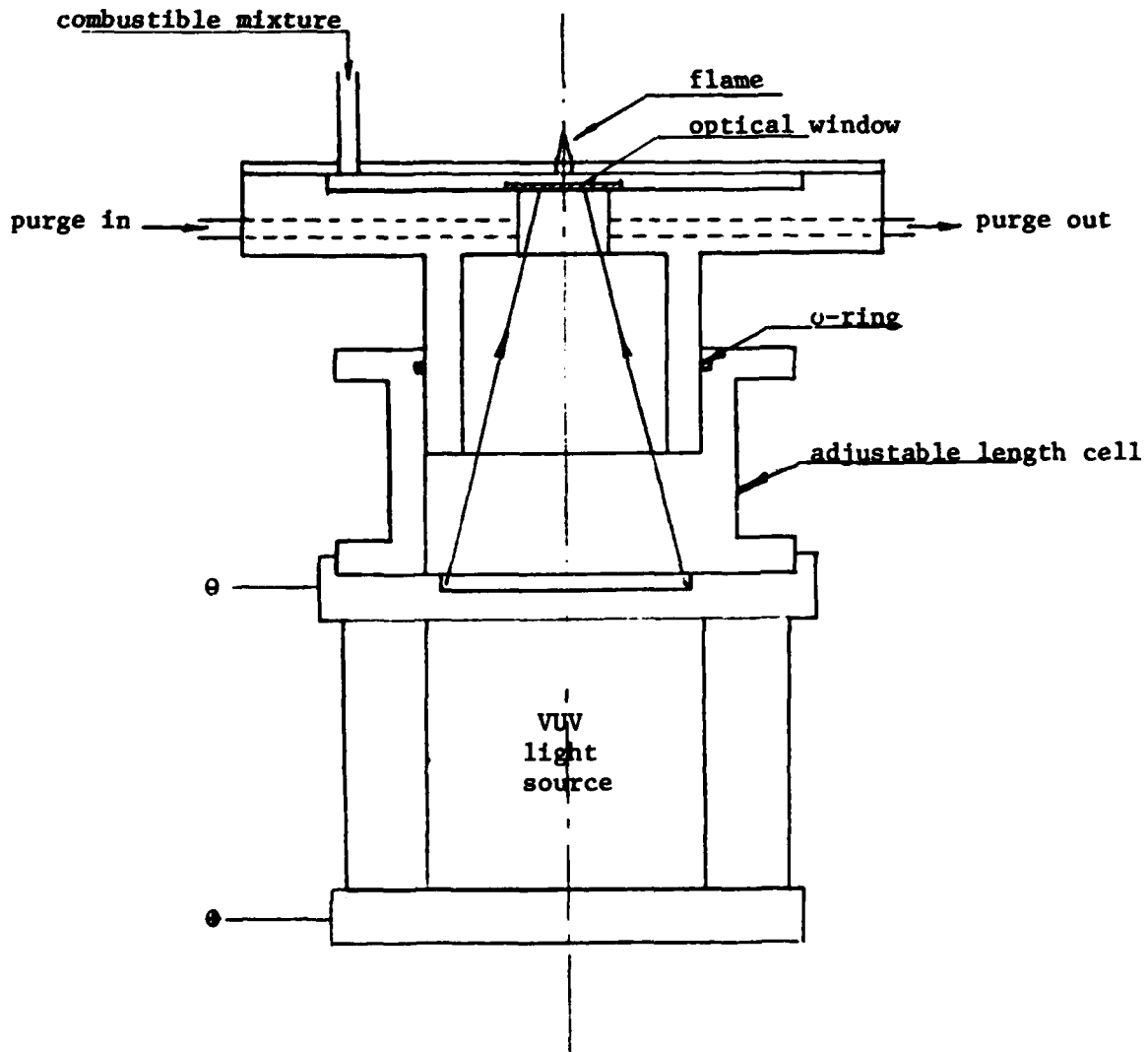


FIGURE 4  
PANCAKE BURNER

Status: The radiative ignition apparatus, and the pancake burner were already fabricated and tested without VUV radiation. Currently we are in the midst of completing the shake down and incorporating the optical components into the system.

The decision made earlier to measure combustion enhancement in terms of changes in burning velocity by detecting flame deflection has been recently supported by Groff and Krage (3) , who successfully measured burning velocity in a similar manner. They reported that flame deflection is indeed a sensitive direct indicator of changes in flame speed. The flame displacement was measured from double-exposure photographs obtained with and without enhancement. We concluded that direct photography in our experiments will suffer from inadequate luminosity, because hydrogen flames are usually too faint in the visible (except at high temperatures and pressures of the detonation wave). Therefore we decided to measure flame deflection by the Schlieren photography. Its chief value appears to be the ability to produce visible contours of density gradients. The differences in density are caused by temperature differences and to a lesser degree by differences in gas composition. The Schlieren system including He-Ne laser was acquired and we are currently experimenting in perfecting it. So far, it was found that the Schlieren method is exceedingly sensitive, and is capable of responding even to very small changes of density generated by hydrogen flames near the lean flammability limit, where we plan to measure combustion enhancement. A detailed description of the Schlieren method is given in Appendix B.

## B. Analytical Effort:

Consistent with the experimental efforts described above, progress in the area of analytical modeling has also proceeded on two fronts: ignition and enhancement.

Ignition: We first reviewed and extended our analytical investigation of photochemical ignition. As a first step, a detailed examination of the kinetics set used in the model was carried out. Specifically, the constants used have been checked against recent compilations of reaction rate data (4). No major differences were found. In some cases, somewhat different preexponential factors and activation energies produced similar overall rate constants when evaluated at temperatures in the initiation range. After the revised values were implemented in our data sets, computer runs revealed either no difference or only slight differences in such ignition measures as fluence required for ignition in laser experiment simulations.

We have also had the opportunity to analyze in greater depth the results of earlier initiation studies. In our previous work, we focused almost entirely on oxygen atom concentration and the input radiant energy (wavelength, irradiance, pulse characteristics) required for ignition. Plots have been prepared of the other radicals generated as a result of the reactions of O and O(<sup>1</sup>D) and of other species in the system. An instructive set of graphs for a case simulating stoichiometric premixed H<sub>2</sub>/O<sub>2</sub> system at 1 atmosphere and 300K, is provided as Figures 5(a,b,c) and 6(a,b,c). The first observation is that all the major free radicals, O, OH, and H, do exhibit similar behavior, as expected. The rapid equilibration of O, OH, and H can be observed by noting the location of the time markers on the graphs. For reference, the VUV irradiation corresponds to a critically damped pulse with a peak at 25 μs. The graphs of the other species exhibit quite different characteristics. The slow radical behavior of HO<sub>2</sub> (metastable) is evident in the less responsive tracking of the pulse than is present in O, OH, and H. The role of H<sub>2</sub>O<sub>2</sub> and O<sub>3</sub> as effective radical sinks is clear from the increase in H<sub>2</sub>O<sub>2</sub> in the two curves for which ignition is not indicated (which actually turn around corresponding to increase in H<sub>2</sub>O<sub>2</sub> concentration), and the virtually constant concentration achieved by ozone in those cases.



FIGURE 5

IGNITION MODELING OF STOICHIOMETRIC  $H_2/O_2$  AT ATMOSPHERIC PRESSURE  
 NUMBERS NEXT TO CURVES ARE RELATIVE RADIATIVE ENERGIES  
 O, H, AND OH PROFILES

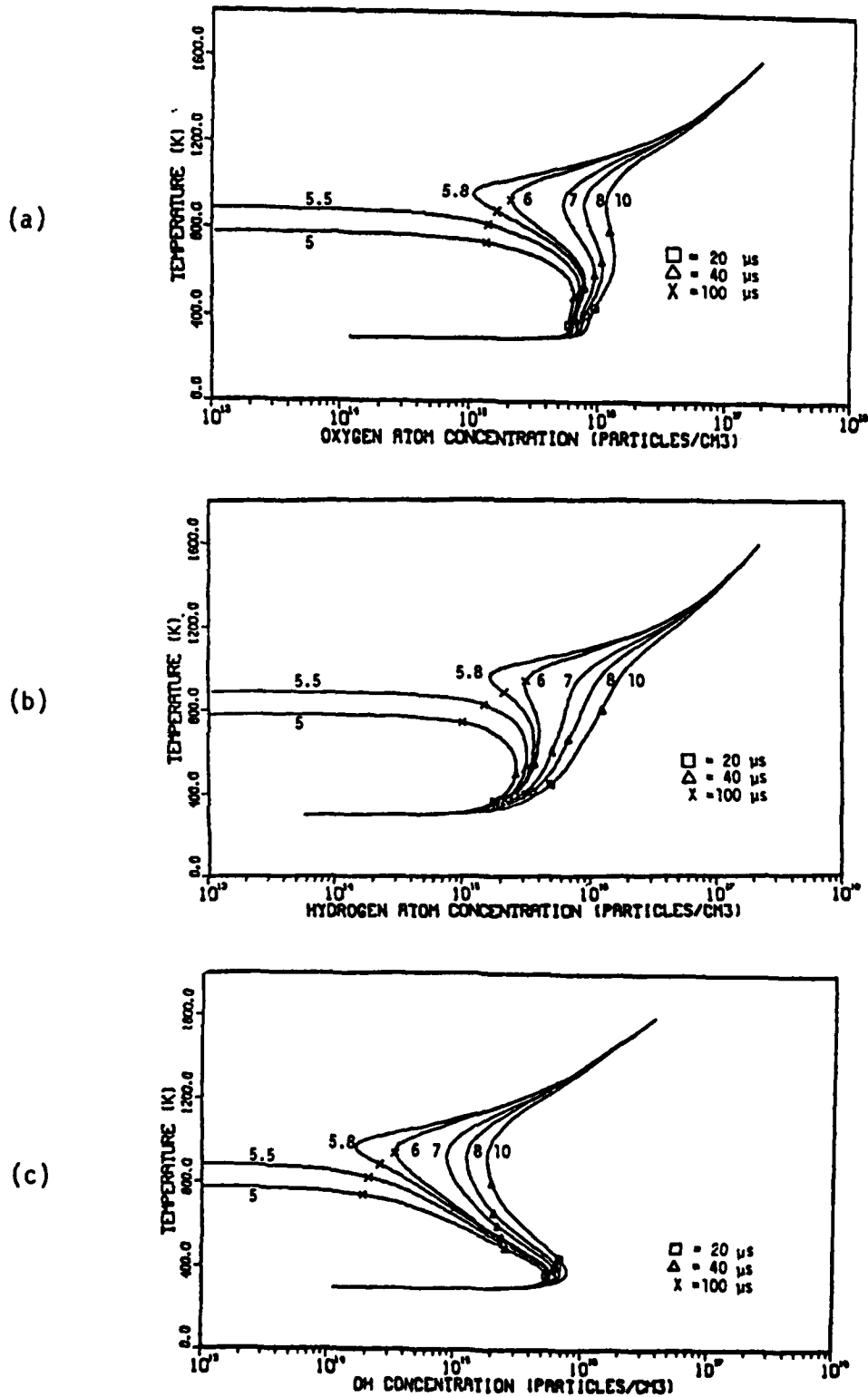
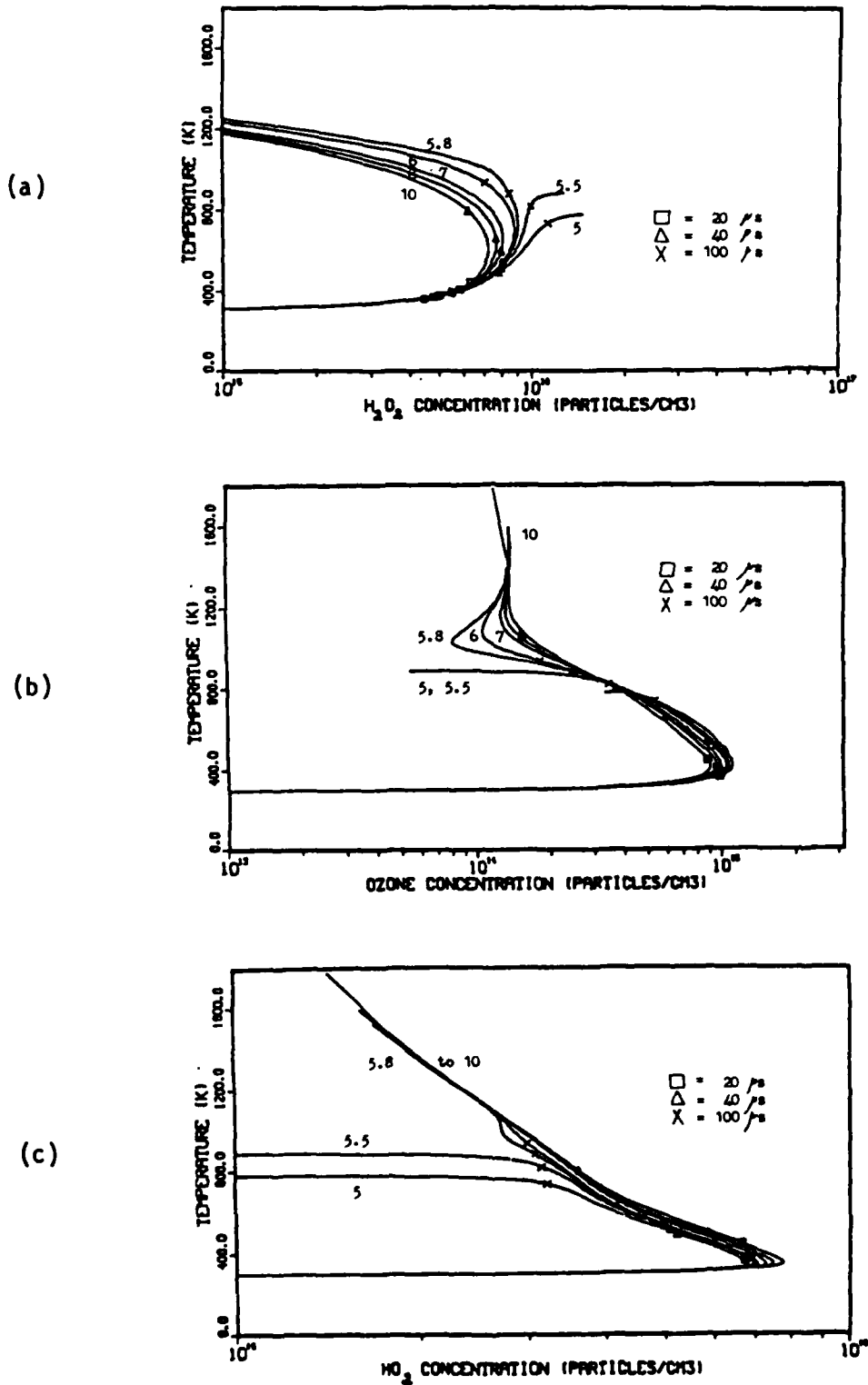


FIGURE 6

IGNITION MODELING OF STOICHIOMETRIC  $H_2/O_2$  AT ATMOSPHERIC PRESSURE  
 NUMBERS NEXT TO CURVES ARE RELATIVE RADIATIVE ENERGIES  
 $H_2O_2$ ,  $O_3$ , AND  $HO_2$  PROFILES



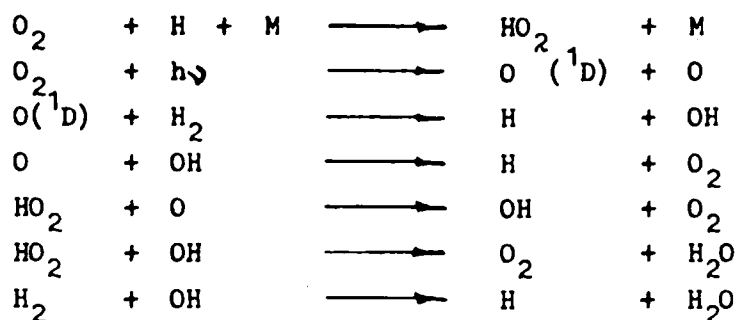
Clearly, chain-terminating reactions of radicals leading to the formation of the stable species  $\text{H}_2\text{O}_2$  and  $\text{O}_3$  are deleterious to combustion initiation. The same observation is true for the formation of the hydroperoxyl radical. Even though it is a free radical and many of its reactions give one of the reactive radicals as a product, it nevertheless represents a chemical "detour" which retards the build-up of the  $\text{O-H-OH}$  radical pool necessary for ignition. The key role of  $\text{HO}_2$  has, of course, been identified by other researchers and pertains in hydrocarbon flames as well, e.g., Westbrook and Dryer (5). As detailed below, these observations have proved valuable for the further investigation and development of radiative combustion augmentation.

The intermediate output from ignition runs have been processed to ascertain which reactions are dominant at various times within laser and arc pulses. Namely, the expressions  $A \exp(-E_a/RT) \prod Y_j^{\nu_j}$  and  $k_{\text{photo}} Y_{\text{absorber}}$  which calculate the chemical and photochemical rates, respectively, were evaluated at the temperature and mixture composition at a specifiable time. The reactions were then ranked according to the resulting magnitudes. A consistent picture emerged for those reactions found to be dominant early in the pulse, (well before peak intensity), approximately at the pulse peak, and toward the tail of the pulse (usually close to the classical "thermal" ignition temperature). We summarize these findings in Table I.

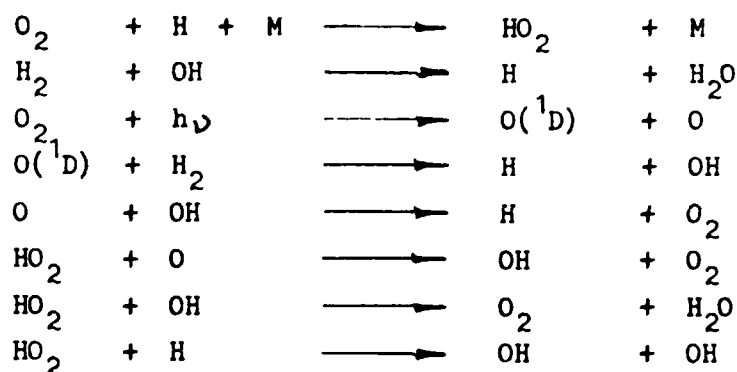
We have also kept abreast of recent, relevant work by other researchers. There is currently considerable interest in combustion ignition and enhancement by radical addition, although few investigators are addressing specific mechanisms for radical production or their implementation in practical combustion systems. Of particular interest is the computational study by T. M. Sloane of General Motors on the effect of selective energy deposition on the homogeneous ignition of methane (6). By suitable choice of initial conditions, the ignition energy was supplied either as heat (pure "thermal" ignition) or as a combination of heat and reactive radicals. In all cases, the total ignition energy, thermal plus dissociative, was kept constant. Induction times were then calculated for methane/air mixtures at various equivalence ratios.

TABLE I  
DOMINATING REACTIONS AT VARIOUS TIMES WITHIN THE LIGHT PULSE

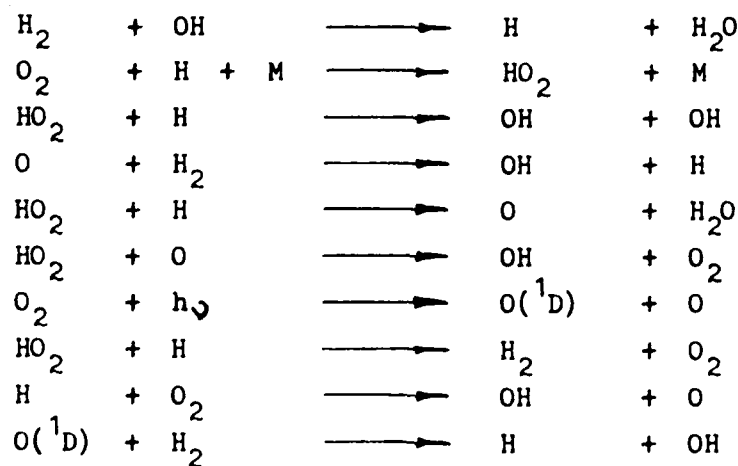
EARLY IN PULSE (TEMPERATURE  $\sim 400^\circ\text{K}$ )



MID - PULSE (TEMPERATURE  $\sim 600^\circ\text{K}$ )



LATE IN PULSE (TEMPERATURE  $\sim 900^\circ\text{K}$ )



Shorter induction times resulted in all cases when a larger fraction of the ignition energy was supplied as reactive radicals. Addition of oxygen atoms gave the shortest induction times in the stoichiometric case; while hydrogen atoms produced the shortest times in the lean case. In both cases, the hydroxyl radical was the least effective. Moreover, the calculations suggested that these shorter induction times may lead to more rapid flame propagation. However, this finding must be regarded as preliminary due to the geometry of the ignition kernel and the neglect of heat and radical transport during the induction period.

Similar modeling studies will be presented by S. Wiriyawit and E. K. Dabora at the 20th International Symposium on Combustion (7). Again, energy supplied as radicals was found to be more efficient for ignition than thermal energy. The mixtures considered were  $H_2$ /air with  $0.3 \leq \phi \leq 1.0$ . In certain instances, an improvement in flame propagation and flame velocity was predicted by their calculations. The results of Sloane and of Wiriyawit and Dabora are encouraging and reconfirm our modeling results and understanding of the role of reactive radicals in radiative augmented combustion.

A preliminary study has been undertaken to determine the adequacy of the HCT computer code (Lund, 8) for radiative ignition modeling. This use would be in addition to its already proven capabilities for calculating flame properties for enhancement as described below.

Enhancement: In the modeling of photochemical enhancement and extension of combustion limits, we have reexamined previous results in greater detail, devised several new cases for study, and planned for future activity. The earlier runs with methane and ethylene as fuels made for us by Dr. Charles Westbrook using the HCT program at Lawrence Livermore Laboratory produced voluminous output which has required much time to review and comprehend. With respect to apparent changes in flame propagation, the results are ambiguous. One problem was the inability within the old HCT set-up to "focus" the photochemical reactions spatially. Thus the photodissociation of molecular oxygen was treated by artificially increasing the rate constant for  $O_2 \rightarrow O+O$  throughout the mixture. At the assumed level of irradiation, this reaction proceeded to produce several effects ahead of the flame front. First,

resulting changes in temperature, and/or pressure in the unburned gas zone can affect the calculated flame propagation velocity to produce anomalous results in some cases. Second, in some runs, a considerable production of partially oxygenated species (e.g., formaldehyde) ahead of the flame was observed. Thus the mixture through which the flame front is proceeding is itself altered in the direction of inhibition. This multiplicity of effects, combined with the complexity of the kinetics for methane and ethylene, makes the earlier results obtained of dubious value.

Before attempting to modify HCT's kinetics subroutine to include spatial photochemical dissociation reactions, we decided to try additional runs for which the kinetics are less complex. The cases chosen for simulation were hydrogen/air flames at stoichiometric and fuel lean (near the flammability limit) conditions at atmospheric pressure. The results of these and later runs are summarized in Table II. Run Number 39 represents the base case for a stoichiometric mixture with no photodissociation. The calculated flame speed of 260 cm/s is somewhat higher than values reported in the literature (9,10). However, at this point we are interested in relative changes in velocities due to UV irradiation and not in absolute values. In Run Number 40, the unimolecular dissociation reaction  $O_2 \rightarrow O+O$  was included with a rate constant of  $3s^{-1}$ , which produced approximately  $10^{13} - 10^{14}$  oxygen atoms/cm<sup>3</sup> ahead of the flame. The calculated flame speed decreased to 250 cm/s. We posited two reasons for this behavior. First, appreciable  $H_2O_2$  was formed ahead of the flame front, resulting in a lower adiabatic flame temperature and tending toward inhibition. This observation is similar to that made for the hydrocarbon flames studied earlier. Second, in a vigorous flame it is likely that at the flame front there is an ample supply of free radicals. In order to observe an effect, we decided to examine cases nearer the lean flammability limit.

The results for an equivalence ratio of 0.6, in fact, showed much the same behavior as the stoichiometric case (Table II, Runs 41, 42). We then proceeded to examine cases with equivalence ratio of 0.4. Run 43 is the base case without photodissociation; Run 44 includes  $O_2 \rightarrow O+O$ , producing approximately  $4 \times 10^{14}$  oxygen atoms/cm<sup>3</sup> ahead of the flame. Again, the

TABLE II

Summary of Enhancement Modeling Runs for H<sub>2</sub>/Air  
at Atmospheric Pressure Using HCT

Run Number	Equivalence Ratio	Simulated Photodissociation Reaction	Rate Constant*	Spatial Extent**	Flame Speed (cm/s)
39	1.0	None	-	-	260
40	1.0	O <sub>2</sub> → O+O	3	R	250
41	0.6	None	-	-	130
42	0.6	O <sub>2</sub> → O+O	3	R	121
43	0.4	None	-	-	47
44	0.4	O <sub>2</sub> → O+O	3	R	43
45	0.4	HO <sub>2</sub> → OH+O	10 <sup>4</sup>	R	60
46	0.4	O <sub>2</sub> → O+O	3	W:700-1000	43
47	0.4	O <sub>2</sub> → O+O	3	W:400- 700	43
48	0.4	O <sub>2</sub> +M → O+O+M	10 <sup>26</sup> x (Nonradiative Value)	W:400- 700	48-50

\* Units of s<sup>-1</sup> for unimolecular reaction

\*\* R = Photodissociation throughout entire reactor

W: T<sub>1</sub> - T<sub>2</sub> = Photodissociation confined to reactor region in which the temperature is between T<sub>1</sub> and T<sub>2</sub> (K).

disappointing result is that of slight inhibition, with the flame speed decreasing from 47 to 43 cm/s. It is instructive to examine the species profiles for these two runs, provided as Figure 7 (a,b,c,d,e). The dashed line in Figure 7a shows the enhanced level of O ahead of the flame for the photodissociative case, but also reveals that just ahead of the front its level drops back virtually to that of the base case. At that same point, the level of  $\text{HO}_2$  is at its maximum (Figure 7d) suggesting that its formation is inhibiting the formation of the O-H-OH pool and working against any possible enhancement due to increased oxygen atom levels. In addition, we observe in Figure 7e that  $\text{H}_2\text{O}_2$  is being formed ahead of the flame.

We postpone discussion of Run 45 and first consider Runs 46 through 48. In an attempt to eliminate the production of  $\text{H}_2\text{O}_2$  ahead of the flame, which may obscure enhancement, we sought to limit the photodissociation of  $\text{O}_2$  to the immediate vicinity of the reaction front. The simplest way of doing so within the HCT structure is to use a temperature window. That is, a temperature interval is specified with the photodissociative rate constant set to the value in Table II when the temperature is within this interval and to a very small value when it is outside. If this interval is suitably chosen, the desired restriction of dissociation to the flame front is achieved. Runs 46 and 47 show the results for two different temperature intervals. Indeed, no  $\text{H}_2\text{O}_2$  was formed ahead of the flame, but there is no change in flame speed from Run 44 which did not show enhancement.

Again, the explanation may be the adequacy of radicals normally generated at the flame front or that the rate constant employed did not produce sufficient oxygen atom concentration at the flame front to give enhancement. Some support for the latter explanation is provided by Run 48. In this run, the photodissociation of  $\text{O}_2$  was modeled by the bimolecular reaction  $\text{O}_2 + \text{M} \rightarrow \text{O} + \text{O} + \text{M}$ . However, the real difference is likely to be that the rate constant used resulted in 25% higher oxygen atom concentration than in Run 44, with the outcome that the flame speed was increased modestly to 48-50 cm/s. We are continuing to investigate the sensitivity of calculated flame speed to the photodissociative rate constant and its relation to the practical delivery and utilization of radiant energy.



FIGURE 7

SPECIES PROFILES FOR RUN NUMBERS 43 AND 44 OF TABLE II  
SOLID CURVES, NO PHOTODISSOCIATION MODELED (RUN 43)  
DASHED CURVES, PHOTODISSOCIATION OF  $O_2$  (RUN 44)  
ABSCISSA IS RELATIVE FLAME POSITION (cm)  
SPECIES INDICATED ON GRAPH

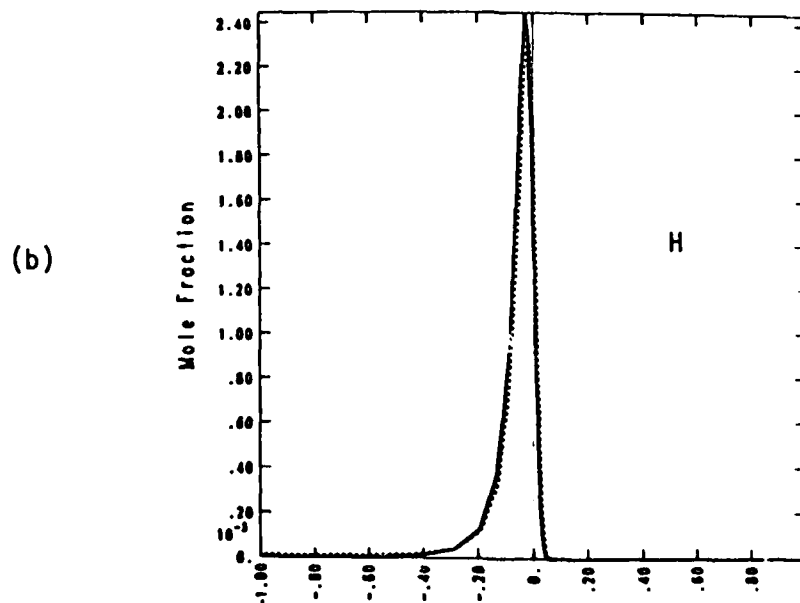
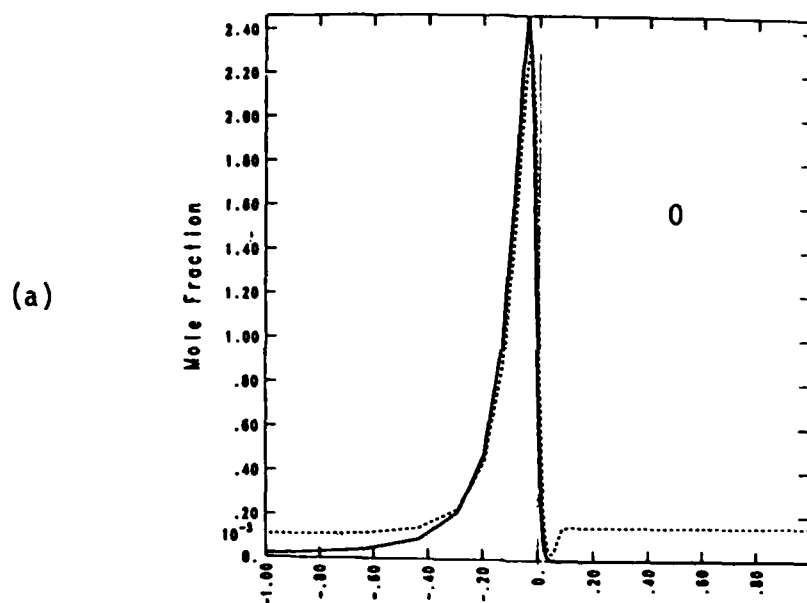
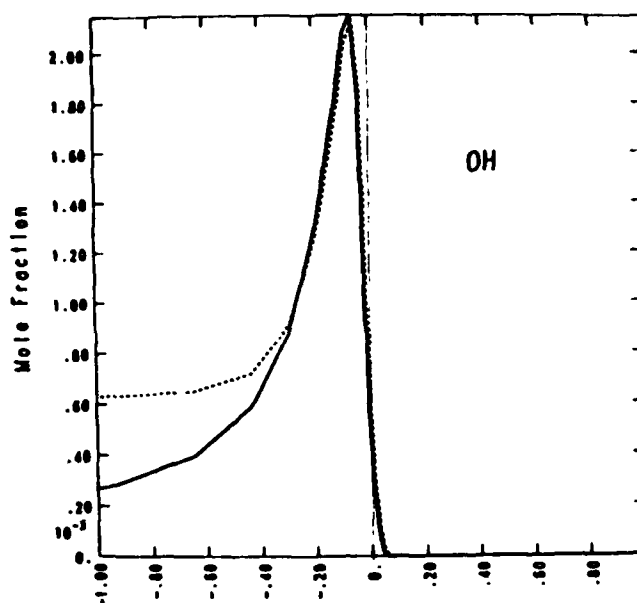
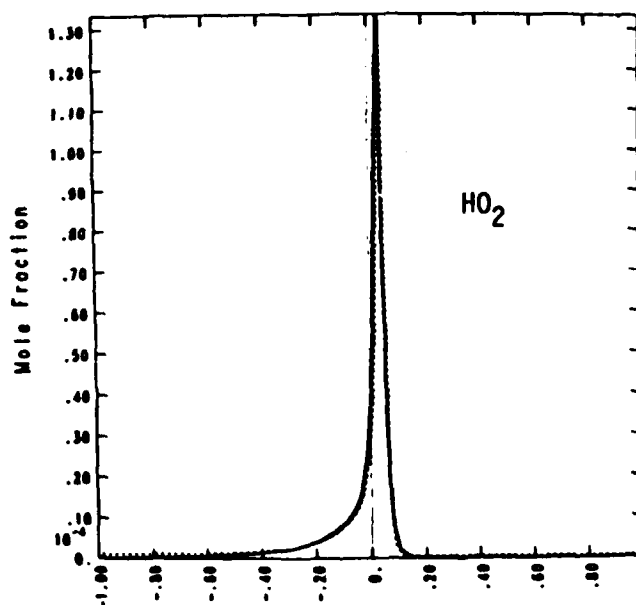


FIGURE 7  
(CONTINUED)

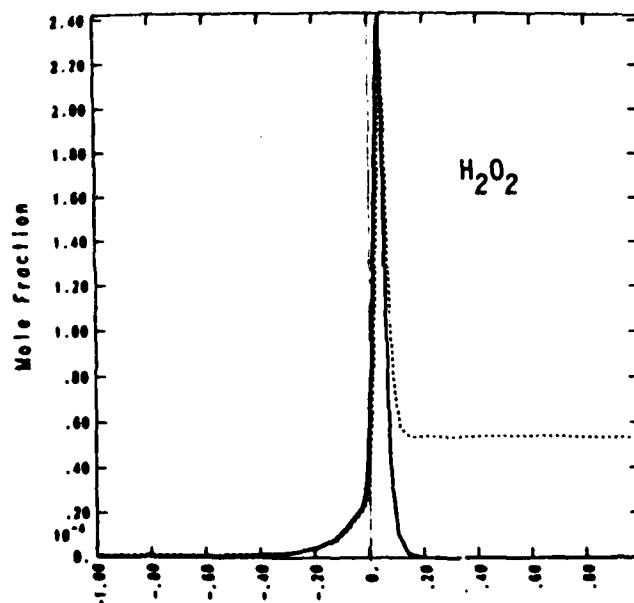
(c)



(d)



(e)



We now turn to the important results of Run Number 45. Upon examination of the wavelength-dependent absorption coefficients for the species in the system, we observed that  $\text{HO}_2$  is an excellent UV absorber. Strong  $\text{HO}_2$  absorption has been observed between 190 nm and 260 nm (11), and weaker absorptions up to 600 nm have also been reported (12). Lack of vibrational fine structure in the absorption spectrum indicates that ultraviolet excitation will promote  $\text{HO}_2$  to a repulsive potential energy curve, causing dissociation. The reaction  $\text{HO}_2 \rightarrow \text{O} + \text{OH}$  was thus incorporated (with no  $\text{O}_2$  dissociation) in Run Number 45 with a rate constant of  $10^4 \text{ s}^{-1}$  estimated on the basis of its absorption cross-section. The result was a considerable enhancement in flame speed as evidenced by a 28% increase to 60 cm/s. The species profiles for the base case and this case are given in Figure 8 (a,b,c,d,e). Examination of these figures reveals a substantial increase in the concentrations of O, H, and OH at the reaction zone and a significant decrease of  $\text{HO}_2$  and  $\text{H}_2\text{O}_2$ . This result points to new practical opportunities for combustion enhancement by allowing more flexibility in possible UV sources (e.g., CW lasers), easier light penetration to the flame zone due to the strong absorption of  $\text{HO}_2$  in regions in which few other species absorb strongly, and relaxed optical requirements (e.g., the use of quartz windows). Clearly, additional cases are suggested to investigate this lead more fully (for example, variation of fuel, equivalence ratio, and intensity of irradiation; inclusion of  $\text{O}_2$  and  $\text{H}_2\text{O}_2$  photo-dissociation). Currently, we are examining these results to determine the most informative runs to be made in the future.

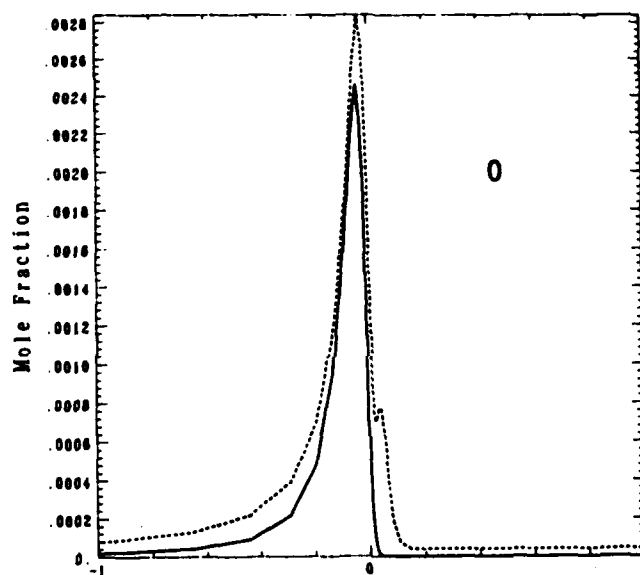
Status: The analytical modeling effort has proceeded on two fronts: ignition and enhancement. After checking the rate constants of our ignition model, a review of our ignition studies of  $\text{H}_2/\text{O}_2$  served to emphasize the role of  $\text{HO}_2$  and identify the dominant reactions. Recent work by several investigators on radical addition in ignition and enhancement has supported our findings and is being followed closely.

The use of the HCT model in a computational study of radiative enhancement has focused on  $\text{H}_2$ /air mixtures at atmospheric pressure. Earlier attempts failed to produce an increase in calculated flame speed with the photodissociation of  $\text{O}_2$  at equivalence ratios from stoichiometric to fuel lean. Unwanted reactions ahead of the flame front and/or low atomic oxygen

**FIGURE 8**

SPECIES PROFILES FOR RUN NUMBERS 43 AND 45 OF TABLE II  
SOLID CURVES, NO PHOTODISSOCIATION MODELED (RUN 43)  
DASHED CURVES, PHOTODISSOCIATION OF  $\text{HO}_2$  (RUN 45)  
ABCISSA IS RELATIVE FLAME POSITION (cm)  
SPECIES INDICATED ON GRAPH

(a)



(b)

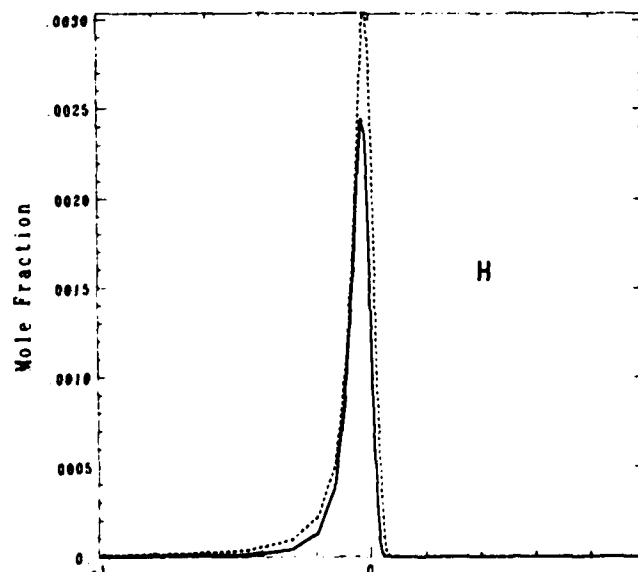
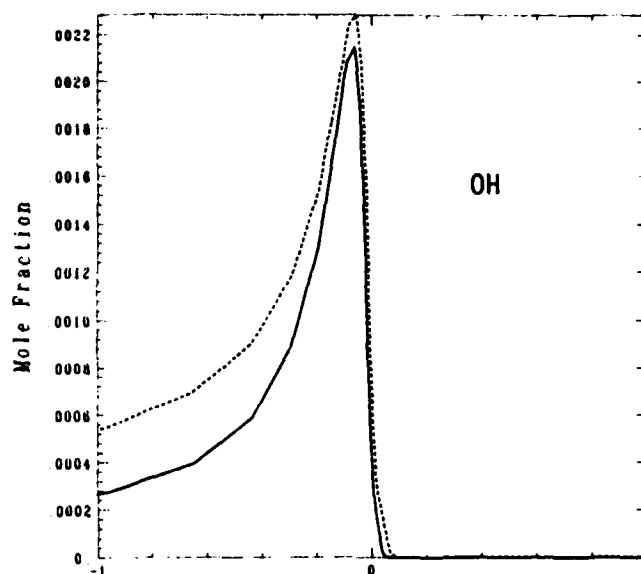
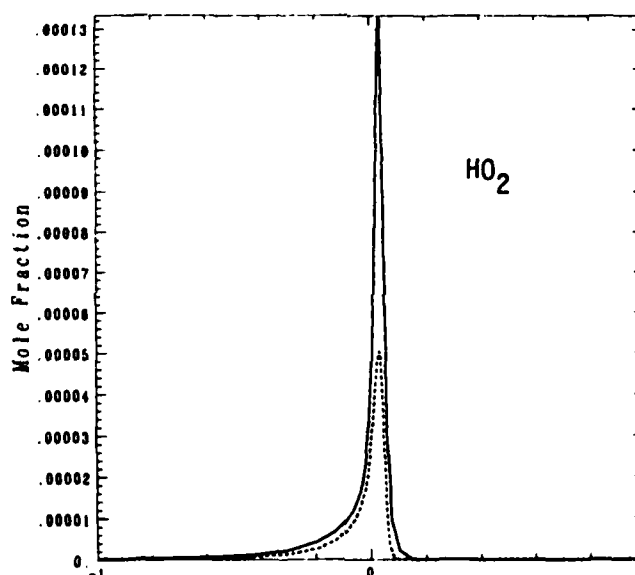


FIGURE 8  
CONTINUED

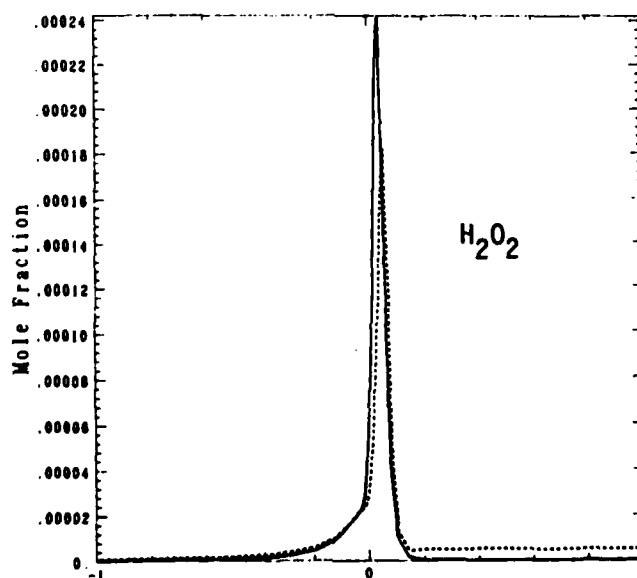
(c)



(d)



(e)



concentration at the flame zone are the probable reasons. We examined these reasons further in the fuel lean case with equivalence ratio of 0.4.

Restriction of the photodissociation to the vicinity of the flame front produced a modest increase in flame speed in one case and a small decrease in two cases with lower concentrations of O. The most significant finding was a 28% increase in flame speed (from 47 to 60 cm/s) with the inclusion of the photodissociation of the metastable hydroperoxyl radical. This result supports the concept of radiative augmentation and points to new practical opportunities for combustion enhancement with relaxed light source and optical requirements. These are being pursued both experimentally and theoretically.

PROFESSIONAL PERSONNEL  
ASSOCIATED WITH RESEARCH EFFORT

Dr. Moshe Lavid - Principal Investigator - PhD Mechanical Engineering,  
State University of New York at Stony Brook, 1974.

Dr. David W. Blair - Senior Staff Member - PhD Mechanical Engineering,  
Columbia University, 1961.

Dr. Arthur T. Poulos - Research Staff Member - PhD Physical Chemistry,  
Northwestern University, 1976.

Dr. J. G. Stevens - Analytical Consultant - PhD Courant Institute of  
Mathematic Sciences, New York University, 1972.

Dr. Charles K. Westbrook - Analytical Consultant - PhD Engineering/Applied  
Science, University of California at Davis, 1974.

### INTERACTIONS

#### A. Presentations

Seven presentations on Radiative Augmented Combustion were made during this reporting period as follows:

- o Baltelle Columbus Laboratories, Columbus, Ohio.
- o Department of Energy, Morgantown Energy Technology Center,  
Morgantown, West Virginia.
- o AFOSR 1983 Contractors Meeting, Scottsdale, Arizona.
- o University of Vermont, CEME Department, Burlington, Vermont.
- o Eastern Section Combustion Institute, 1983 Technical Meeting,  
Providence, Rhode Island.
- o Drexel University MEM Department, Philadelphia, Pennsylvania.
- o AFOSR 1984 Contractors Meeting, Pittsburgh, Pennsylvania.

#### B. Interest Expressed by Other Researchers

Interest in our Radiative Augmented Combustion is reflected by the many requests for additional technical information listed below. Special interest was expressed by researchers working on plasma jet ignition and flameholding such as Professor Felix Weinberg at Imperial College, London, Professor Eli Dabora at the University of Connecticut and Dr. G. Burton Northam at NASA Langley, Hampton, Virginia. NASA's interest in our radiative technique has been pursued to a cooperative experiment in which ENERGIA provides the VUV light sources to be mounted on NASA supersonic combustion facility. The objective is to determine the feasibility of utilizing VUV photo-dissociation for ignition and flameholding in supersonic combustion (scramjets).

The interest in the whole subject of combustion enhancement is currently expanding. Ward and Wu (13) proposed microwave radiation as a means of enhancing the flame speed to allow use of lean mixtures in spark-ignition engines. According to their theory, the microwave field principally heats the electrons of the flame, and the hot electrons transfer energy to the reacting species. The resulting increase in the population of higher vibrational states of the reacting species, especially that of oxygen, is said to increase reaction rates in the flame, as suggested by Jagers and von Engel (14). This increase in the reaction rates is expected to be greater than that which results from simple thermal heating of the bulk gases. Ward (15,16) compared theoretically and



experimental cavity quality factors and concluded that the microwave flame-plasma interaction was strong as predicted. Additional work by Ward (17) in a constant-volume combustion bomb suggests that burn time can be reduced by the application of microwaves in conjunction with plasma-jet ignition. Subsequent experiments by Clements and coworkers (18,19) show that the flame-plasma electrons are heated and that changes in flame speed are on the order of 5% to 10% when gas breakdown does not occur. Recently, several researchers (3) at General Motors Research Laboratory reported an increase of 6% in burning velocity of fuel-lean premixed laminar flames with microwave radiation. However, they concluded that the enhancement is not due to a radiative (photochemical) effect but rather due to a thermal effect. The increase in flame speed is explained in terms of simple microwave heating of the bulk gases in the flame zone, which yields a higher flame temperature. ENERGIA has established open communication with Drs. T. Sloane, R. Hickling and E. Groff from the General Motors group.

During this period the following professionals expressed interest in our research:

Prof. Eli K. Dabora, University of Connecticut, Storrs, Connecticut.  
 Dr. F. E. Fendell, TRW, Redondo Beach, California.  
 Dr. Edward G. Groff, GM Research Laboratories, Warren, Michigan.  
 Dr. Alan Hartford, Los Alamos Scientific Lab, New Mexico.  
 Prof. Clarke E. Hermance, University of Vermont, Burlington, Vermont.  
 Dr. Lyle O. Hoppie, Eaton Corporation, Southfield, Michigan.  
 Dr. David Mann, US Army Research Office, Research Triangle Park, North Carolina.  
 Dr. G. Burton Northam, NASA, Langley Research Center, Hampton, Virginia.  
 Prof. Herschel Rabitz, Princeton University, Princeton, New Jersey.  
 Dr. Tom Sloane, GM Research Laboratories, Warren, Michigan.  
 Prof. Felix Weinbert, Imperial College, London, England.  
 Dr. Charles K. Westbrook Lawrence Livermore Lab, University of California.

SUMMARY: ASSESSMENT OF RESEARCH RESULTS

The radiative technique has demonstrated the capability to both ignite and enhance combustion processes and to broaden normally encountered stability limits.

The research on radiative ignition and combustion enhancement is providing fundamental information on a unique combustion process. Concepts which represent a new departure and extension of conventional combustion practice can evolve from the experimental data being obtained. Aspects of the radiative ignition and enhancement concept have been already demonstrated in the laboratory under static (no flow) conditions. Successful pulsed light source ignition experiments reconfirm the radiative augmented concept and demonstrate the technical feasibility of designing an advanced optical-radiative igniter. Reproducible ignitions with a continuous light source imply the possibility of using the light as an optical radiative flame stabilizer with no pressure loss instead of the conventional intrusive flameholders. Encouraging results of radiative combustion enhancement in terms of higher flame propagation velocities and larger extinction times and distances suggest a potential to extend the combustor operating limits. It is based on knowledge that the increase in flame propagation is indicative of higher combustion rate which can result in extension of the operating envelope of flash-back and blow-off limits.

From the encouraging analytical results reported here we gain confidence that radiative augmented combustion can become a potentially viable technique for extending current aircraft operating limits associated with combustion phenomena. However, supportive experimental results under flow conditions are required before eventual application to gas turbine engine systems can be envisioned both for improved combustor operation and flameholding. Some future areas of potential application are: high altitude combustor reignition following flame-out, drag-free flame stabilization in supersonic combustor, and added flexibility for conventional combustors to use future alternate fuels. To this end, radiative ignition and combustion enhancement experiments under flow conditions have been designed, and are planned to be conducted during the second year of this program, together with a continued VUV light source development in the direction of improved beam optics.

APPENDIX ACalculation of Concentration of Photochemically Produced Radicals

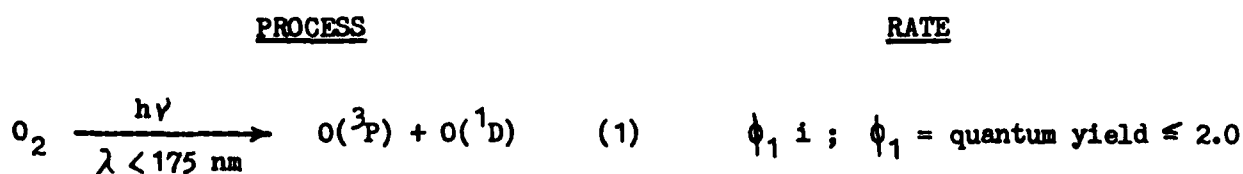
It is possible to estimate what experimental conditions are required in order to produce a detectable change in burning velocity. This can be accomplished because burning velocity is a strong function of the concentration of radicals which participate in chain branching reactions. In the case of radiative augmented combustion, increases in steady-state radical concentration can, in turn, be related to the photon absorption rate. The photon absorption rate is determined by the experimental conditions, e.g., VUV intensity incident on the reactor window, the excitation frequencies (which influence absorption rate through the wavelength dependence of the absorption coefficient), the pathlength through which the VUV beam must pass to reach the reaction zone, the dimensions of the reaction zone, and the concentrations of the absorbing species.

First, we shall calculate the concentration of O which must be produced photochemically to achieve a detectable change in burning velocity. If the rate of change of burning velocity with change in (O) were known, one could calculate the concentration of O required for a desired amount of enhancement. This rate data was not available for O, but only for H. However, calculations have shown (20) that the concentrations of H and O required to sustain a flame are on the same order of magnitude. Therefore, we shall use data which relates H to burning velocity, and assume that burning velocity varies with O in a similar way.

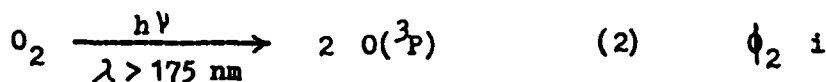
In a  $H_2 - O_2 - N_2$  mixture having burning velocity 200 cm/sec, the rate of change of burning velocity with change in (H) can be estimated from literature data (21) as  $2.9 \times 10^{-14} \frac{\text{cm}}{\text{sec}} / \frac{\text{particles}}{\text{cm}^3}$ . For a 20 cm/sec increase in burning velocity (easily detectable in a differential experiment, i.e., velocity with radiation on, compared to radiation off), an additional  $6.9 \times 10^{14}$  radical particles/cm<sup>3</sup> must be deposited in or near the reaction zone.

Secondly, we will derive a relationship between (O) and  $i$ , the photon absorption rate (photons absorbed per cm<sup>3</sup> per sec). From the simplified

reaction scheme (1) - (4), the steady-state  $O(^3P)$  concentration during irradiation with  $\lambda < 175 \text{ nm}$  can be approximated as (5):



$i = \text{photons absorbed}$   
per  $\text{cm}^3$  per sec



(3) & (4) — chain propagation

$$(O(^3P)) = \frac{\phi_1 i}{k_3 (H_2)} \quad (5)$$

Examining equation (5):

$\phi_1$ , the quantum yield for formation of  $O(^3P)$  from  $O_2$ , is expected to be approximately 2.0, based upon the quantum yield for  $O_3$  formation in  $H_2 - O_2$  mixture at 1 atm following irradiation at 147 nm (22).

$k_3$  is strongly temperature dependent ( $k_3 = 4.1 \times 10^{11} e^{-4950/T}$ , where  $T$  is in Kelvin (3)).

$(H_2)$  is, of course, a function of the gas pressure, temperature, and its composition. Values for  $(H_2)$  can be calculated assuming the ideal gas law and a mole fraction of 0.143, corresponding to stoichiometric conditions.

If the reaction zone is approximated as flat, an expression for  $i$ , the photons absorbed in the reaction zone per  $\text{cm}^3$  per second, is easily derived. Consider a parallel, continuous beam of VUV entering a cylindrical burner as shown schematically in Figure A1.

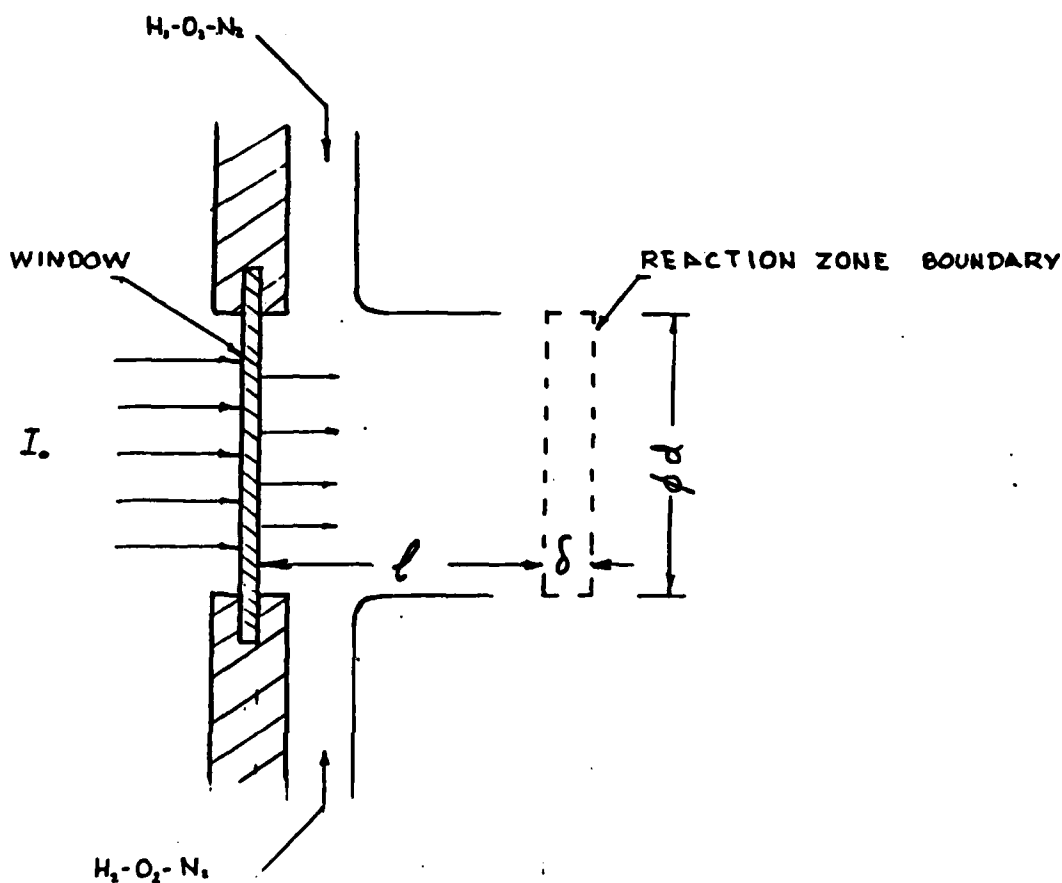


FIGURE A1

CONFIGURATION OF THE PANCAKE BURNER

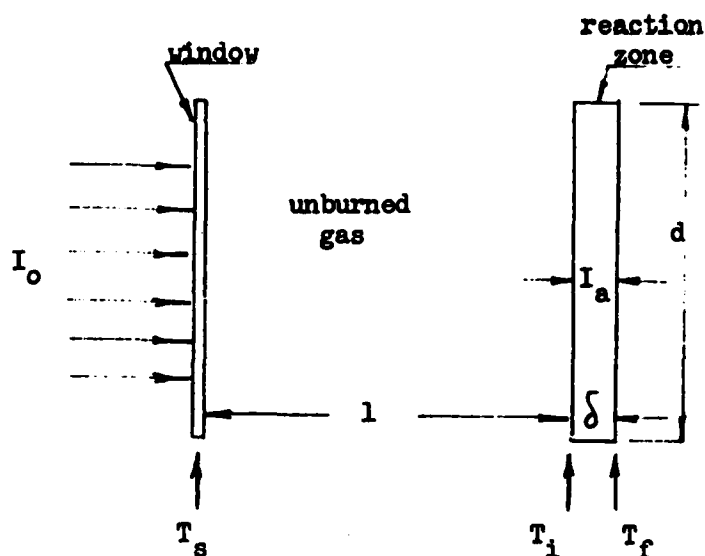
Let  $I_o$  = photon flux incident on the sapphire window, photons/sec

$T_s$  = transmittance of sapphire window

$T_i$  = transmittance of gas at distance  $l$

$T_f$  = transmittance of gas at distance  $(l + \delta)$

$I_a$  = photons absorbed per second, in the reaction zone.



$$\text{Then } I_a = T_s I_o (T_i - T_f). \quad (6)$$

By the Beer-Lambert relationship,

$$T_i = e^{-Kpl} \quad (7a)$$

$$T_f = e^{-Kp(l + \delta)} \quad (7b)$$

Where  $K$  is the absorption coefficient in  $\text{atm}^{-1} \text{cm}^{-1}$   
and  $p$  is the pressure of absorbing gas.

The reaction cylinder has volume  $\delta \pi (d/2)^2$ , so the number of photons absorbed per  $\text{cm}^3$  per sec,  $i$ , becomes

$$i = \frac{T_s I_o (e^{-Kpl} - e^{-Kp(l + \delta)})}{\delta \pi (d/2)^2}$$

$$1 = \frac{T_s I_0 (10^{-K_{p1}/2.303}) (1 - 10^{-K_p \delta / 2.303})}{\int \pi (d/2)^2} \quad (8)$$

Here only absorption by  $O_2$  is considered because  $K_{H_2} = 0$  and  $K_{N_2} < 0.1 \text{ atm}^{-1} \text{ cm}^{-1}$ , while  $K_{O_2} = 8.5 \sim 396 \text{ atm}^{-1} \text{ cm}^{-1}$  in the VUV region,  $\lambda < 175 \text{ nm}$  (23). Absorption by  $H_2O$  has been ignored because its absorption coefficient is approximately five times less than that for  $O_2$ , and also because absorption interferes with that of  $O_2$  only in the thin reaction zone where it is generated.

$I_0$  can be measured by gaseous chemical actinometry. A variety of chemical systems have been employed by others (24).

An estimation of  $I_0$  must be made rather than measuring it because the apparatus for enhancement tests is currently being designed. We use reported spectral irradiance for an EIMAC lamp. For a 300 W lamp, an average of .015 W/nm radiant power has been measured (25). This value can be increased to .025 W/nm because the lamp to be used in these experiments is 500 W. The range of predominant photon absorption by  $O_2$  is 140 - 170 nm, so one obtains a value for  $I_0$  of 0.75 watt. If the energy appeared only as photons with  $\lambda = 156 \text{ nm}$  (a wavelength in the "middle range" of oxygen absorption), then a conversion factor of  $7.8 \times 10^{17}$  photons/J is obtained. Using the sapphire transmittance at 156 nm of .722,  $T_s I_0$  is calculated to be  $4.2 \times 10^{17}$  photons/sec.

We are now in a position to calculate the concentration of O atoms produced by irradiation, using equations (5) and (8) for a given set of experimental conditions. The calculation will be illustrated with one example, in which the parameters are reasonably close to the pancake burner described in the Research Progress section.

Substituting in equation (8),

$$1 = 0.5 \text{ cm}, d = 0.5 \text{ cm}, \delta = 0.1 \text{ cm},$$

and,

$$K = 182 \text{ atm}^{-1} \text{ cm}^{-1}, \lambda = 156 \text{ nm}, p_{O_2} = 7.6 \text{ torr (total pressure = 53.2 torr, stoichiometric H}_2 \text{ - air ratio),}$$

and

$$T_s I_0 = 4.2 \times 10^{17} \text{ photons/sec,}$$

yields,

$$i = 1.1 \times 10^{18} \text{ photons cm}^{-3} \text{ sec}^{-1}.$$

At this pressure and a temperature of  $600^\circ\text{K}$ ,

$$(H_2) = 1.2 \times 10^{17} \text{ particles cm}^{-3},$$

$$k_3 = 1.07 \times 10^{-14} \text{ cm}^3 \text{ sec}^{-1} \text{ particle}^{-1}.$$

Substituting  $i$ ,  $(H_2)$ ,  $k_3$  and  $\phi_1 = 2.0$  into equation (5),

results in,

$$(O) = 1.7 \times 10^{15} \text{ particles/cm}^3.$$

We thus conclude that under these conditions, steady state irradiation produces a sufficiently high concentration of oxygen atoms at a temperature of  $600^\circ\text{K}$  to produce an unambiguously detectable change in burning velocity.

It is desirable to determine what experimental conditions will maximize the concentration of  $O$  produced by irradiation. By substituting equation (8),  $k_3$ ,  $\phi_1$ , and  $(H_2)$  (assuming ideal gas law) into equation (5), we get the following expression for  $(O)$ ,



$$(0) = \frac{2.0 T_s I_o (10^{-K P_{O_2}} l^{1/2.303}) (1 - 10^{-K P_{O_2} \delta / 2.303})}{4.1 \times 10^{11} \exp\{(-4950/T)\} \left(\frac{P_{H_2}}{RT}\right) \pi \delta (d/2)^2} \text{ particles/cm}^3 \quad (9)$$

Where  $P_{H_2}$  = pressure of  $H_2$ , atm

$P_{O_2}$  = pressure of  $O_2$ , atm

$R$  = the gas constant, .0821 l - atm/mole  $^{\circ}K$

$T$  = temperature,  $^{\circ}K$

Equation (9) can be differentiated and equated to zero in order to determine what are reasonable experimental variables which yield maximum  $O$  concentration in the reaction zone.

The effect of four of the variables,  $K$ ,  $l$ ,  $\delta$ , and  $P_{O_2}$  on (0) can be illustrated by examining a new quantity EXPT. EXPT is that part of equation (9) which contains those variables.

$$EXPT = (10^{-K P_{O_2}} l^{1/2.303}) (1 - 10^{-K P_{O_2} \delta / 2.303}) / \delta \quad (10)$$

Values for  $\delta$ ,  $l$ , and  $K$  were chosen and  $P_{O_2}$  was incrementally varied until EXPT reached a maximum. The values of  $P_{O_2}$  and the total pressure,  $P_T$ , at  $EXPT_{max}$  are listed in Table A1.

Examining these values, one notices that  $EXPT_{max}$  always decreases as  $l$  is increased. This effect is the justification for the pancake

TABLE A1

## PRESSURES RESULTING IN MAXIMUM (O) AT VARIOUS GEOMETRICAL PARAMETERS AND WAVELENGTHS

VUV at 157 nm ( $K = 167 \text{ atm}^{-1} \text{ cm}^{-1}$ ) ( $F_2$  laser)

$l$ , cm	$\delta$ , cm	$P_{O_2}$ (torr)	$P_{\text{total}}$ (torr)	EXPT <sub>max</sub>
.2	.1	18.6	130.3	1.48
.5	.1	8.36	58.5	.670
1	.1	4.18	29.3	.350
2	.1	2.28	16.0	.179
5	.1	.76	5.3	.072
.5	.01	9.12	63.8	.728
.5	.05	8.74	61.2	.700
.5	.15	7.9	55.9	.642
1	.05	4.56	31.9	.359
1	.2	4.18	29.3	.335
1	.5	3.80	26.6	.296

VUV at 173 nm ( $K = 14.1 \text{ atm}^{-1} \text{ cm}^{-1}$ )

.5	.1	98.8	691.6	.670
1	.1	51.3	359.1	.350
2	.1	26.6	186.2	.179
5	.1	10.6	74.5	.073
10	.1	5.3	37.2	.037

VUV at 193 nm ( $K = 0.027 \text{ atm}^{-1} \text{ cm}^{-1}$ ) ( $ArF$  laser)

1	.1	$2.66 \times 10^4$	$1.86 \times 10^5$	.350
50	.1	$5.70 \times 10^2$	$3.99 \times 10^4$	$7.31 \times 10^{-3}$

burner design, in which the distance between the radiation source and the flame front is made small ( $\sim .5$  cm).

The effect of pressure is also noteworthy. At wavelengths of 157 - 173 nm, and  $l = .5$  cm, EXPT is optimized at a pressure of less than one atmosphere. Thus, the pancake burner should be operated at reduced pressure or with mixtures that are highly diluted with nitrogen. At 193 nm, the low value of  $K$  causes EXPT to be optimized only at very high pressure. This fact can be used to rationalize the failure to obtain ignition by ArF laser pulses at atmospheric pressure (26).

A final consideration is the temperature dependence of  $k_3$  in equation (9). At high reaction temperatures (e.g., 2380 °K for  $H_2 - O_2 - N_2$ , 1 atm, stoichiometric)  $k_3$  becomes so large as to reduce (0) below the value targeted to produce enhancement. However, we believe that in the preheat zone and also in lower temperature regions ( $T < 700$  °K) of the reaction zone, (0) will be sufficiently large to produce enhancement.

In summary, we have calculated that the concentration of O required to produce detectable change in burning velocity is on the order of  $10^{15}$  particle/cm<sup>3</sup>. Moreover, the current EIMAC lamp with the configuration of the pancake burner seems to be capable of producing adequate atomic oxygen. Finally, the design also allows for some degree of flexibility in selecting the geometrical parameters of the apparatus as well as the total pressure of the gas mixture. This, in turn, will permit a further increase in the concentration of O, if deemed necessary to achieve detectable enhancement in burning velocity.

## APPENDIX B

## MEASUREMENT OF FLAME DEFLECTION BY SCHLIEREN METHOD

It was concluded that radiative augmented combustion can be best determined by measuring increase in burning velocity (see Status of the Research-Enhancement). Burning velocity is the velocity relative to the unburnt gas, with which a plane, one-dimensional flame front travels along the normal to its surface. It is the eigenvalue of the 1-D flame equations, and is a fundamental (physico-chemical) constant for a given combustible mixture. Unfortunately, although its theoretical concept is quite simple, its practical measurement is very difficult. Consequently, there is no agreement on a standardized method, and conflicting values of burning velocities continue to be published. We decided earlier (20) to avoid the non-stationary flames because of the additional complexity in the incorporation of the VUV radiation to a moving flame, and to use stationary flames. For stationary flames, there are four different types of burner and several measurement techniques for measuring burning velocity; (1) open circular tube and nozzle, (2) flat flame burner (3) slot burner and (4) direct measurement of unburnt gas velocity. We selected the first burner, and already fabricated a burner called "pancake" which produces a conical flame. Conical flames have been used by the majority of researchers, and the derivation of the burning velocity is given below.

Figure 1B shows an idealized flame, which is assumed to be conical. The dashed line represents the surface  $a_0$ , that is the locus of all points where the gas temperature rises just above the initial temperature  $T_0$ . The solid line is the optically visualized surface of the flame  $a_v$ , which may be either a

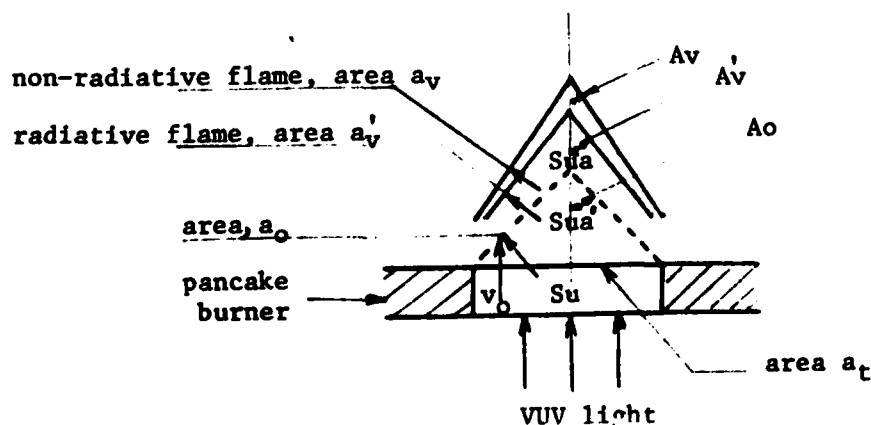


FIGURE 1B

Conical Flame Above the Pancake Burner

luminous, shadow, or Schlieren surface (to be discussed later). The cross-sectional area of the burner is  $a_t$ , and the mean velocity of the premixed gas in the burner is  $v_o$ , which is easily measured by a flow meter. The burning velocity  $S_u$  is given by equation (1B), from conservation of mass.

$$S_u = \frac{v_o a_t}{a_o} \quad (1B)$$

or, in terms of the cone apex angle,

$$S_u = v_o \sin A_o \quad (2B)$$

Difficulties arise from the facts that the flame is not perfectly conical, the burning velocity is not constant, and the surface area  $a_o$  cannot be optically visualized. Instead researchers measure the apparent burning velocity  $S_{ua}$ , which is referred to the optically visualized surface  $a_v$ .

$$S_{ua} = \frac{v_o a_t}{a_v} \quad (3B)$$

or, in terms of the cone apex angle,

$$S_{ua} = v_o \sin A_v \quad (4B)$$

From equations (1B) and (3B) we obtain,

$$\frac{S_u}{S_{ua}} = \frac{a_v}{a_o} \quad (5B)$$

The area ratios for different visualized surfaces; schlieren, shadow and direct photography are reported (27) as 1.11, 1.14 and 1.21 respectively. These values show that all visualized surfaces tend to underestimate the true burning velocity, and that the schlieren and shadow surfaces are better than the luminous (direct photography) surface. However, there are considerable difficulties in the optical interpretation of shadowgraph records, that can lead to serious errors in burning velocity. In addition, hydrogen flames selected for our experiments are usually too faint in the visible for direct photography. This left us with only one choice, the schlieren method.

The chief value of schlieren photography appears to be its ability to produce visible contours of density differences. It is exceedingly sensitive, being capable of responding even to very small changes of density at relatively low temperatures typical of hydrogen flames. The principle of this method is based on the fact that the change in refractive index is inversely proportional to the absolute temperature. The unburnt gases are heated on their way to the reaction zone of the flame. The increase in temperature causes changes in their density and refractive index. The change in refractive index is used to determine the position of the reaction zone. The schlieren line is observed at that part of the flame where the gases begin to be preheated. Several optical arrangements (9, 28) can be used for the schlieren method. Our optical layout is shown in Figure 2B.

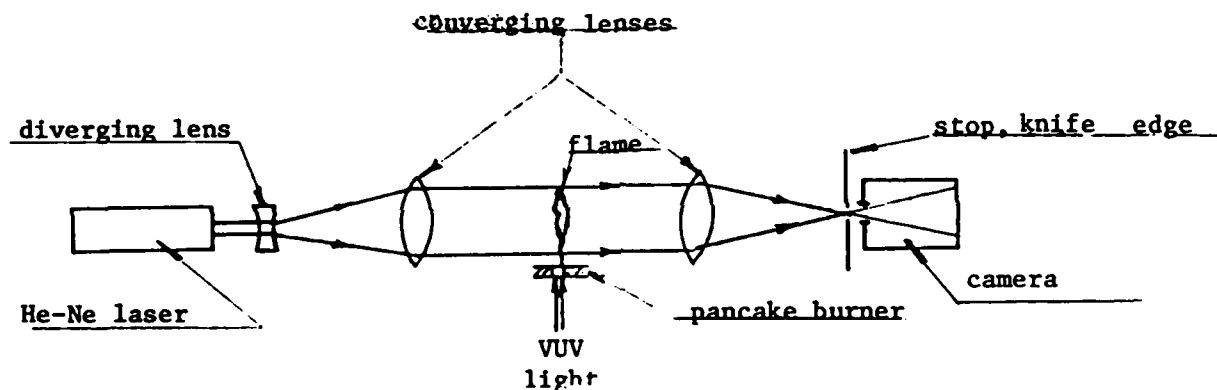


Figure 2B - Schlieren Optical Arrangement

This is a modification (29) of a typical arrangement. Here, a He-Ne laser is used as the light source because of its superior optical purity. Inserting a He-Ne laser-line filter in front of the camera prevents scattered light from the VUV source from entering the camera. Also, rays distortions by lens achromaticity is eliminated. Once the schlieren lines are photographed, a relative value for the visualized surface  $a_v$  can be easily obtained by graphical integration on the a projection of the negative. The relative value does not cause a problem, as long as the same camera lens - setting is used for the measurement of the burning velocity under radiative and non-radiative conditions. The effect of VUV radiation is manifested by a change in flame position. An increase in burning velocity is partially balanced by a deflection upstream, to a region of higher

local velocity. This provides the required additional stabilization closer to the burner. This upstream displacement of the flame changes the visualized surface from  $a_v$  to  $a_v'$ . Accordingly, equation (3B) can be written for the radiative case as,

$$\frac{Sua'}{Sua} = \frac{v_o a_t}{a_v'} \quad (6B)$$

and

$$\frac{Sua'}{Sua} = \frac{a_v}{a_v'} \quad (7B)$$

Rather than measuring the relative visualized surface areas under non-radiative and radiative conditions,  $a_v$  and  $a_v'$ , we can measure the cone apex angles  $A_v$  and  $A_v'$ , respectively. In this case, the increase in flame speed is expressed by equation (8B),

$$\frac{Sua'}{Sua} = \frac{\sin A_v'}{\sin A_v} \quad (8B)$$

Another way to measure the change in burning velocity was recently suggested by Groff and Krage. (3) After a flame deflection is obtained as a result of radiative enhancement, the light is switched off, and an attempt is made to produce the same flame deflection without radiation by increasing the mass flow rate of the combustible mixture. This incremental increase in flow rate is more directly related to an absolute change in burning velocity. However, it may require fine and tedious flow adjustments.

The decision to conduct differential experiments discussed in section "Status of the Research", circumvents the major problem of being unable to measure  $a_o$  with accuracy, and the difficulty associated with having conflicting values of burning velocity in the literature. Our differential measurements will indicate the increase in the apparent flame speed, thus eliminating the need for having  $a_o$  and  $Su$ . A comparison of the values of the apparent burning velocity with and without VUV radiation will determine the level of combustion enhancement. These values can be corrected to yield the increase in true burning velocity by using equation (5B) provided, of course,  $a_o$  is either known or a correction factor is derived.

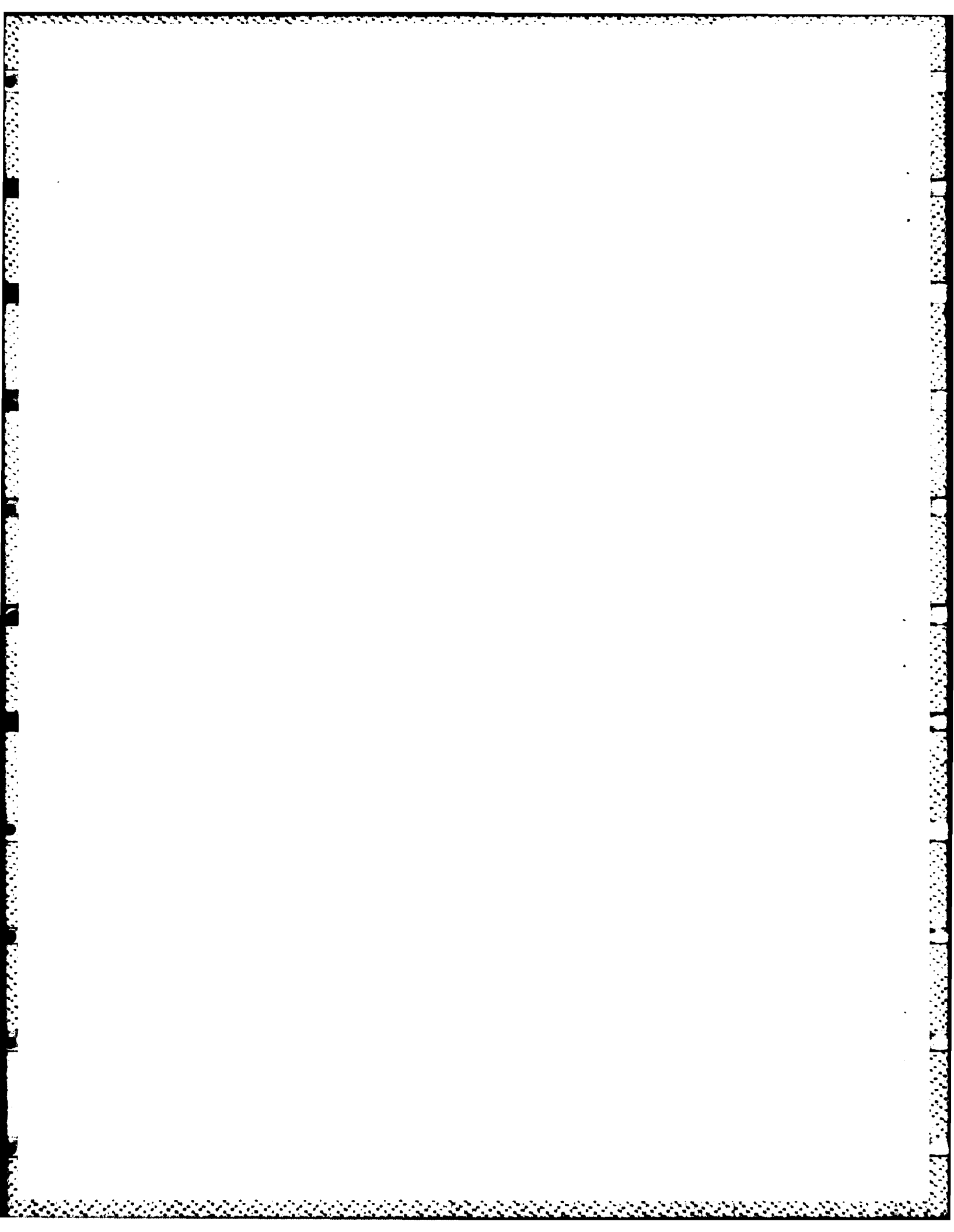
In summary, flame deflection seems to be a very sensitive method for measuring even small changes in burning velocity, and schlieren method appears to be the most suitable one for our experiments.

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